

STRESS-DEPENDENT ELECTRICAL CONDUCTION IN GRANULAR MATERIALS

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A thesis submitted to fulfil requirements for the degree of Doctor of Philosophy

Statement of Originaltiy

This is to certify that to the best of my knowledge, the content of this thesis is my own work.
This thesis has not been submitted for any degree or other purposes.

I certify that the intellectual content of this thesis is the product of my own work and that all
the assistance received in preparing this thesis and sources have been acknowledged.



Chongpu Zhai

19/09/2017

Abstract

This dissertation is focused on electrical conduction behaviour in granular systems with the purpose of acquiring a fundamental understanding towards applications of granular materials. The electrical properties of granular materials are of great significance in many engineering applications, such as powder metallurgy processes, optimization of battery electrodes, piezo-resistive composites for tactile sensing, and soil water content measurements. Performance in such systems can be largely influenced by complex multi-physics interactions arising from microstructures of granular materials. The bulk of this dissertation is built on six published or submitted papers. Each chapter is prefaced by an introductory section presenting the motivation for the corresponding paper and its context within the greater body of work.

After project background and related previous work introduced in Chapter 1 and 2, respectively, Chapter 3 and 4 deal primarily with the contact properties between rough surfaces. The obtained information at the interfacial scale serves as an experimental and numerical basis for modelling inter-particle contacts in granular media. Specifically, in Chapter 3, the first and second papers show the effects of roughness and fractality on the normal contact stiffness of rough surfaces using experimental and numerical approaches, respectively. In Chapter 4 containing the third and fourth papers, we extend discussions to the correlation of mechanical and electrical properties with scaling analyses, based on experimentally measured normal contact stiffness and electrical contact resistance.

Chapter 5 with the fifth paper presents the effects of network configuration on macroscopic network responses focussing on the dielectric universal scaling behaviour. A unified description has been obtained for the emergent scaling properties of network responses for random two-phase systems with varying topological configurations. The established network is pivotal for connecting the effective behaviour at the surface scale to macro scale responses of granular materials.

In Chapter 6, the final paper shows a physical picture illustrating experimentally observed alternating-current universal scaling in conductive granular systems under different stress states. An effective numerical approach incorporating inter-particle interaction has been provided to simulate electrical responses of granular materials. The combination of the studies from macro-scale phenomena, network topologies, and inter-particle properties is presented leading to new physics-based constitutive models that contain lower scale information.

This dissertation presents a new comprehensive understanding of conduction behaviour in granular materials by means of a physics-based framework combining features containing both experimental and numerical information obtained across various length scales, guiding design and optimisation of various granular materials.

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Publications and Awards

The following publications have been produced as a result of this Ph.D. study:

Journal papers:

1. Zhai, C., Gan, Y., Hanaor, D., Proust, G., & Retraint, D. (2016). The role of surface structure in normal contact stiffness. *Experimental Mechanics* 56 (3), 359-368.
2. Zhai, C., Hanaor, D., Proust, G., & Gan, Y. (2017). Contact stiffness of multiscale surfaces by truncation analysis. *International Journal of Mechanical Sciences* 131, 305-316.
3. Zhai, C., Hanaor, D., Proust, G., & Gan, Y. (2015). Stress-dependent electrical contact resistance at fractal rough surfaces. *Journal of Engineering Mechanics* 143(3).
4. Zhai, C., Hanaor, D., Proust, G., Brassart, L., & Gan, Y. (2016). Interfacial electro-mechanical behaviour at rough surfaces. *Extreme Mechanics Letters* 9, 422-429.
5. Zhai, C., Hanaor, D., & Gan, Y. (2017). Universality of the emergent scaling in finite random binary percolation networks. *PLoS ONE* 12 (2).
6. Zhai, C., Hanaor, D., Proust, G., & Gan, Y. (2017). Stress-dependent electrical transport and its universal scaling in granular materials (in submission).

Conference papers:

1. Zhai, C., Hanaor, D., Proust, G., & Gan, Y. (2017) Electrical transport in granular metals. *The 8th International Conference on Micromechanics of Granular Media (P&G2017, 3-7 Jul)*, Montpellier, France.
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3. Zhai, C., Gan, Y., & Hanaor, D. Numerical analysis of normal contact stiffness of rough surfaces. (2015) *The 2nd Australasian Conference on Computational Mechanics (ACCM2015, Nov 30-Dec 1)*, Brisbane, Australia.
4. Zhai, C., Bevand, S., Gan, Y., Hanaor, D., Proust, G., Guelorget, B., & Retraint, D. (2014) Measurement of normal contact stiffness of fractal rough surfaces. *The 23rd Australasian Conference on the Mechanics of Structures and Materials (ACMSM2014, 9-12 Dec)*, Byron Bay, Australia.

The following awards have been received during the course of this thesis.

1. Paul and Dee-Dee Slade Young Investigator Award, 2016, IEEE Holm, Unites States;
2. Postgraduate Research Support Scheme, 2016, The University of Sydney;
3. Best Student Poster, Research Student Conference, 2015, The University of Sydney;
4. Annie B Wilson Prize, 2014, The University of Sydney;
5. Endeavour Postgraduate Scholarship: Prime Minister Awards, 2014-2017, Australia.

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To whom it may concern,

as the coauthor of for the following publications, I confirm that:

(1) Chongpu Zhai was the first author of the journal paper “Stress-Dependent Electrical Contact Resistance at Fractal Rough Surfaces”, published in Journal of Engineering Mechanics in 2015, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.

(2) Chongpu Zhai was the first author of the journal paper “The Role of Surface Structure in Normal Contact Stiffness”, published in Experimental Mechanics in 2016, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.

(3) Chongpu Zhai was the first author of the journal paper “Interfacial electro-mechanical behaviour at rough surfaces”, published in Extreme Mechanics Letters in 2016, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.

(4) Chongpu Zhai was the first author of the journal paper “Contact stiffness of multiscale surfaces by truncation analysis”, published in International Journal of Mechanical Sciences in 2017, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.

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(6) Chongpu Zhai was also the first author of the manuscript “Stress-dependent electrical transport and its universal scaling in granular materials”, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.



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With this letter I hereby confirm:

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Chongpu Zhai was also the author of the manuscript “Stress-dependent electrical transport and its universal scaling in granular materials”, as part of his Ph.D. For this work, Chongpu lead the research effort and wrote the paper himself.

Sincerely,

A handwritten signature in blue ink, consisting of stylized, overlapping loops and a long horizontal stroke extending to the right.

Dorian Hanaor



THE UNIVERSITY OF
SYDNEY

Gwénaëlle Proust

Associate Professor, School of Civil Engineering

31 August 2017

To whom it may concern,

I confirm:

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1 Introduction

Granular materials are ubiquitous not only in the real nature but also in a variety of engineering applications. The electrical conduction of granular materials combines the unique properties of individual granules and collective properties arising from interconnections between granules. In this chapter, the background of electrical conduction in granular materials and related research are introduced.

1.1 Granular materials

Granular materials are conglomeration of macroscopically discrete solid particles, which are large enough such that they are not subject to thermal fluctuations. The lower size limit for grains in granular materials can be as small as nanoscale. On the upper size limit, the physics of granular materials may be applied to ice floes where the individual grains are icebergs and to asteroid belts of the Solar System with individual grains being asteroids (Duran, 2012).

Granular materials are commercially important in applications as diverse as pharmaceutical industry, agriculture, and energy production (Duran, 2012). According to material scientist Patrick Richard, "Granular materials are ubiquitous in nature and are the second-most manipulated material (Patrick et al., 2005) in industry (the first one is water)".

Research into granular materials can be traced back at least to Charles-Augustin de Coulomb, whose law of friction was originally stated for interactions between grains (Duran, 2012). The evolution of the particles follows Newton's equations, with repulsive forces between particles that are non-zero only when there is a contact between particles. Although granular materials are very simple to describe they exhibit a tremendous amount of complex behaviour, much of which has not yet been satisfactorily explained. They behave differently than solids, liquids, and gases which has led many to characterize granular materials as a distinct form of matter. In some sense, granular materials do not constitute a single phase of matter but have characteristics reminiscent of solids, liquids, or gases depending on the average energy per grain. However, in each of these states granular materials also exhibit properties which are unique. Granular materials also exhibit a wide range of pattern forming behaviour when excited, e.g., vibrated or allowed to flow, injected heat and current. As such granular materials under excitation can be thought of as an example of a complex system (Duran, 2012; Herrmann et al., 2013).

1.2 Electrical conduction in granular materials

The discovery and characterization of electromagnetic waves by Heinrich Hertz at the end of the nineteenth century ignited studies of the interactions of these new waves with various types of materials. Among them, the electrical (and heat) transport in conductive granular materials presents unique properties and have found numerous applications in industry, such as electronic components (Beloborodov et al., 2007; Dyre et al., 2009), combustion reactors (Maniatis et al., 2001), fusion

reactors (Gan et al., 2010), batteries (Ott, 2015), piezo-resistive composites for tactile sensing (Stassi et al., 2014), metallurgy processes and 3D printing (Arifin et al., 2014) etc. The transport behaviour of granular materials is contributed by not only the grains themselves, such as their bulk electrical resistivity, elasticity, plasticity, and surface structure, but also the way they are arranged in the packing featured by the density, network topology, and state of compaction.

The granularity brings about new physics over a wide range of length scales from macro down to nano and atom scales, further extending the already rich list of remarkable effects exhibited by disordered systems (Beloborodov et al., 2007; Mehta, 2007). Recent years have seen remarkable progress in the development of micro and nano technologies which allow the design and fabrication of granular conductors at smaller length scales (Beloborodov et al., 2007; Mowbray and Skolnick, 2005; Murray et al., 2000). By altering the size and shape of granules one can regulate the inter-particle interaction, e.g., quantum tunnelling effects (Beloborodov et al., 2007). In particular, by tuning the microscopic parameters, one can create granular materials varying from relatively good conductors to pronounced insulators, controlled by the material properties of the grains, inter-particle interaction, degree of disorder and network topography (Beloborodov et al., 2003). This makes granular conductors a new class of artificial materials with tuneable electronic properties, therefore, a perfect exemplary system for studying the metal-insulator transition and related phenomena, with implications for engineering applications.

Many efforts have been made to understand electrical conductivity of granular materials composed of conductive and insulating constituents. The intensive research in this field is motivated not only by their important technological promises but by the appeal of dealing with an experimentally accessible model system that is governed by tuneable cooperative effects of disorder, electron correlations, and network configuration (Beloborodov et al., 2007; Beloborodov et al., 2003; Mowbray and Skolnick, 2005).

1.3 Phase transitions of electrical conduction

1.3.1 Branly effect

One of the first major findings of electrical conduction in granular materials is the phase transition observed by Édouard Branly in 1890 (Branly, 1892). Metal fillings in a non-conductive container tend to present a normal state of high electrical resistance due to the naturally existing of oxide layer. In the presence of an electrical potential difference or receiving the transient electromagnetic waves generated by an electrical spark at some distance, the initially high electrical resistance drops down by several orders of magnitude, becoming practically conductors (Dilhac, 2009; Hirlimann, 2007; Zhai et al., 2015). It has been early observed that a permanent and unique percolation path is along with the drop in resistance of the granular medium (Dorbolo et al., 2003), leading to the understanding that welding of the grains was the result of the coherer effect. Because of this welding of the grains the effect is not reversible although the electrical path can be easily broken by gently striking the fillings, restoring the original large resistance of the granular medium (Vandembroucq et al., 1997). Additionally, mechanical pressure allows better contacts between grains that reduce the original resistance, reducing the sensitivity to the external excitation (Zhai et al., 2016a; Zhai et al., 2015).

This effect was incorporated in practical radio detectors insuring the pioneering developments of radio-telegraphy (Hirlimann, 2013; Kryzhanovskii, 1992) including the first wireless radio

transmission. During the last decades, interests for the Branly effect resumed (Creysseels et al., 2007; Creysseels et al., 2008, 2009; Dorbolo et al., 2007; Falcon and Castaing, 1993; Falcon and Castaing, 2005; Falcon et al., 2004), focusing on the electrical breakdown of oxide layers on grains (Dorbolo et al., 2007; Falcon and Castaing, 2005; Houssa et al., 1998; Vandembroucq et al., 1997), the modified tunnelling effect through the metal-oxide/semiconductor-metal junction (Holm, 1967), the attraction of grains by molecular or electrostatic forces (Dorbolo et al., 2003; Dorbolo et al., 2007), and local welding of micro-contacts by a Joule heating effect (Béquin and Tournat, 2010; Dorbolo et al., 2002, 2003; Dorbolo et al., 2007). A global process of percolation within the grain assembly also has been investigated (Vandembroucq et al., 1997). Among the phenomena proposed to explain the coherer effect, it is easy to show that some have only a secondary contribution. Particularly, the coherer effect was observed with a chain of conductive grains, i.e., one-dimensional structure, at a sufficiently high imposed voltage, in a way similar to the action at a distance of a spark or an electromagnetic wave (Falcon and Castaing, 2005), demonstrating that the Branly effect is primarily arising from the inter-particle contact rather than percolation mechanism with in the whole granular assembly.

1.3.2 Frequency-dependent electrical responses

The other important aspect of electrical conduction in granular materials is the frequency-dependent electrical responses, from DC (direct current) to AC (alternating current) regimes. The determination of AC conductivity, σ , and permittivity, ϵ , of granular materials is a complicated problem that depends on many parameters: the statistical distribution of the shape and size of the grains, the applied force, and the local properties at the contact scale of two grains, that is, the degree of oxidization and surface roughness (Béquin and Tournat, 2010; Bourbatache et al., 2012; Creysseels et al., 2007; Dyre et al., 2009; Falcon et al., 2004; Zhai et al., 2016b).

Conductive granular materials consisting of randomly distributed conducting and insulating phases can be considered as disordered dielectric systems, such as ceramic composites (Almond and Bowen, 2004; Li and Schwartz, 2006), ion/electron-conducting glasses (Almond and Bowen, 2004; Dyre et al., 2009; Schrøder and Dyre, 2008), amorphous semiconductors (Dyre et al., 2009; Elliott, 1987), etc. In these systems, various parameters govern the electrical, thermal, chemical, and/or mechanical properties of components of a system across multiple scales from molecular up to macroscopic length-scale. Experimental and computational research efforts are increasingly conducted in order to gain insights into how these properties combine across scales to determine overall system performance. Particularly, the AC behaviour of these mentioned systems have been extensively studied. A similar conductivity-frequency dependence was proposed by Jonscher (1977b) and is known as the “Universal Dielectric Response” (Jonscher, 1977a; Zhai et al., 2017). This emergent property does not arise directly from any particular physical or chemical properties of the involved components, but rather is a consequence of the way components combine (Almond et al., 2013; Almond et al., 2011; Creysseels et al., 2008; Mccullen et al., 2009; Murphy et al., 2006). More importantly, the conductivity spectra for ionic and electronic conduction in crystalline and amorphous systems (Bakkali et al., 2016; Dyre et al., 2009; Roling, 1998) under a wide range of temperature conditions demonstrate a single master curve, suggesting the validity of the time-temperature superposition principle.

These dielectric mixtures have been effectively approximated as a random network of resistors and capacitors (Almond and Bowen, 2004; Creysseels et al., 2008; Mccullen et al., 2009) with representative conductors exhibiting a constant conductance and dielectric components exhibiting a variable complex admittance which is directly proportional to an angular frequency. As well as being

important in understanding the electrical properties of granular materials, the approach based on resistor-capacitor network provides a useful test bed for identifying different types of emergent behaviour, finding the critical parameters associated with the observed network responses, determining fundamental drives, and addressing the question regarding the universality of complex system behaviour.

1.4 Research objectives

This dissertation is focused on conduction behaviour in granular materials with the purpose of acquiring a fundamental understanding towards applications of granular energy materials. In granular materials, the granules are large enough to possess distinct electronic characteristics compared with the overall granule aggregates, but sufficiently small to exhibit network responses in mesoscopic scale. A constitutive model unifying disparate systems at various length scales for the electrical conduction in granular materials is of considerable importance. This work describes the conduction behaviour of granular materials by introducing a physics-based framework combining features containing experimental and numerical information obtained across various length scales, thus gaining access to a comprehensive model that can be used directly for granular system design and optimisation.

1.5 Thesis structure

The project background and related previous work are introduced in Chapter 1 and 2, respectively, followed with six published or submitted papers as the bulk of this dissertation. Each paper is prefaced by an introductory section presenting the motivation for the corresponding paper. The first four papers are concentrated on the rough interfacial properties in Chapter 3 and 4. The obtained information at surface scale can serve as the experimental and numerical basis for modelling particle contact in granular media. The following fifth paper in Chapter 5 presents the influences of the configurations of fabricated networks of granular materials by interactions between granules on network electrical responses. The established network is pivotal for connecting the effective behaviour at the surface scale to parameters of macro scale of granular materials. In the last paper in Chapter 6, the combination of the studies from macro phenomena, network topologies, and inter-particle properties are presented leading to new physics-based constitutive models that contain lower scale information. Finally, Chapter 7 concludes this work by summarising the findings and implications, and provides an outlook on future work in contact mechanics and granular physics.

2 Related Work

In this chapter, related work on the electrical conduction of granular materials is reviewed. More detailed and topical literature review with more details can be found in following chapters.

2.1 Effective properties of granular media

The problem of electrical conduction in heterogeneous materials including granular materials is mathematically analogous to the problems of thermal conductivity, permittivity and magnetic permeability. The study of these topics dates back to early works of James Clerk Maxwell and Lord Rayleigh in the late 19th century (Carson et al., 2005; Dyre and Schröder, 2000). Since then, studies of heterogeneous dielectric materials have been in a continuous state of development. The heterogeneous structure is replaced with a hypothetical material which is homogeneous one-component material with equivalent conductivity, diffusivity, coefficient of thermal expansion, mechanical properties etc. Numerous models have been proposed in the last century for the estimation of effective properties, e.g., elastic properties, thermal properties, electrical and fluid transport properties of composite or porous materials. Detailed information can be found in the reviews of (Carson et al., 2005; Clerc et al., 1990; Landauer, 1978; Pietrak and Wisniewski, 2015; Torquato, 2013; Wang and Pan, 2008). Regarding the approaches, it can be distinguished between averaging methods derived analytically for simplified microstructures (such as effective medium theory and percolation theory), or Monte-Carlo methods, determined based on numerically generated microstructures.

Widely used for the estimation of effective transport properties are the effective medium approximations (EMA), which belong to the class of mean-field theories. They are based on the mathematical solution for the disturbance of a homogeneous field due to a spherical inclusion as for example derived by Maxwell (Maxwell, 1881). Due to their nature, effective medium approximations are unable to accurately predict the properties of heterogeneous materials beyond the percolation threshold. Maxwell was the first to give analytical expressions for effective conductivity of heterogeneous media in his famous work on electricity and magnetism. He considered the problem of dilute dispersion of spherical particles of conductivity k_p embedded in a continuous matrix of conductivity k_m , where thermal interactions between filler particles were ignored (Bird et al., 2007). Maxwell's expression is as follows:

$$\frac{k_{\text{eff}}}{k_m} = 1 + \frac{3\varphi}{\left(\frac{k_p + 2k_m}{k_p - k_m}\right) - \varphi}, \quad (2.1)$$

where φ is the volume fraction of the filler. Based on this model, Rayleigh (Rayleigh, 1892) considered a material in the form of spherical inclusions arranged in a simple cubic array, embedded in a continuous matrix. Hamilton and Crosser (1962) included effects of different particle shapes. Hasselman and Johnson (1987) emphasized that the interfacial properties between filler and matrix tend to have a large influence on the overall conduction behaviour thus should be taken into account. Base on Eq. (2.1), different approximations can be derived to estimate the effective conductivity of

composite materials. Widely used models are the so-called self-consistent approach and differential approximation. For the self-consistent approach, the composite at a certain arbitrary mixing ratio is considered as a homogenized matrix phase with an effective conductivity (Carson et al., 2005). In the second approach, the composite material is assumed to be constructed incrementally by introducing infinitesimal changes to an already existing material, leading to differential equations (Bruggeman, 1935). However, the approximation based on the aforementioned approaches yields unsatisfactory results for compositions with widely different conductivities, predicting a spurious percolation threshold independent of microstructure (Ott, 2015). In general, effective medium approximations fail to predict the properties of multiphase media close to their percolation threshold. Recent work (Ambrosetti et al., 2010; Bertei and Nicoletta, 2011; Pennetta et al., 2002) further incorporated percolation phenomenon, which will be discussed in the following section.

2.2 Percolation theory

Percolation theory was first developed by Flory (1941) and Stockmayer (1943) to describe how small branching molecules react and form very large macro-molecules (Sahimi, 1994). Percolation phenomena arise in a vast field of applications: transport behaviour, mechanical properties, glass transition, spread of fire and diseases, etc. (Aharony and Stauffer, 2003). The classical percolation theory centres on the occurring sharp transition for site and bond percolation network. This transition point is called the percolation threshold which is fundamentally important in estimating the effective conductivity in multi-phase medium. When increasing the amount of conductive phase, one eventually reaches a point, i.e., percolation threshold, at which chains of conductive components begin to appear. Formation of such conductive channels causes a significant increase in the effective conductivity of the medium. This effect is visible as a shift from a flat to a steep slope of the effective conductivity plotted versus the volume fraction of conductive phase.

2.2.1 Conductivity near the percolation threshold

For the description of the electrical percolation, we consider a mixture of a metallic component having a conductivity, σ_R , and a dielectric component with conductivity, σ_C , corresponding to the occupied sites/bonds and non-occupied sites/bonds, respectively. Particularly, σ_C demonstrates an increasing trend with increasing frequency, approaching infinity. We further consider the scenarios at low-frequency region ($\omega \rightarrow 0$) and high-frequency region ($\omega \rightarrow \infty$) by incorporating conductor-capacitor mixture and superconductor-conductor mixture (Aharony and Stauffer, 2003; Sahimi, 1994)

In the case of a conductor-capacitor mixture we have $\sigma_C=0$. As the percolation threshold, P_T , is approached from above ($P_R > P_T$), the effective conductivity, σ_1 , for low-frequency scenarios follow:

$$\sigma_1 \propto \sigma_R (P_R - P_T)^\mu, \quad (2.2)$$

where μ is a positive constant depending on the dimensionality of the network. While the volume fraction of conductive phase, P_R , below the percolation threshold, we have no conductivity, as shown in the Fig. 1

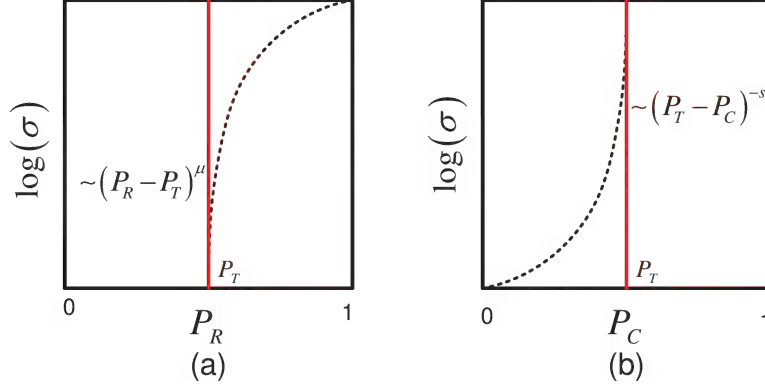


Fig.1. Schematics of the percolation for conductor-capacitor mixture at (a) low-frequency, and (b) high-frequency.

In the other case of a superconductor-conductor mixture where $\sigma_C \rightarrow \infty$ with infinitely high frequency. As the percolation threshold is approached from below ($P_c < P_T$), the effective conductivity σ_h at high frequency can be described:

$$\sigma_h \propto \sigma_C (P_T - P_c)^{-s}, \quad (2.3)$$

with the conductivity becoming infinite above the percolation threshold (Aharony and Stauffer, 2003; Sahimi, 1994). Here s is a positive constant depending on the dimensionality of the network.

2.2.2 Finite-size scaling

The value of the percolation threshold is mathematically defined for an infinitely large system (Sahimi, 1994). Percolation in finite systems deserves discussion because practical applications usually involve finite systems (Clerc et al., 1990). According to the finite-size scaling theory, the effect of finite size of the system can be shown through the effective values of percolation threshold $P_T(L)$ for a finite system of linear size L :

$$P_T = P_T(L) \propto L^{-\frac{1}{\nu}}, \quad (2.4)$$

where the value of ν depends on the system dimensionality (Sahimi, 1994). The finite size of the network cause a shift in the percolation threshold.

2.2.3 Percolation in binary granular mixtures

The concept of coordination numbers is central to the practical implementation of percolation theory (Chen et al., 2009; Gan and Kamlah, 2010; Gan et al., 2010; Ott et al., 2013; Zhai et al., 2017). Broadly speaking, the coordination number represents the number of contacts a particular particle makes with its neighbouring particles. For a mixture of conductive and non-conductive grains randomly packed, each grain can be regarded as a lattice site and the contacts between spheres are bonds. In percolation theory, it is distinguished between occupied and unoccupied sites. This correlated to conducting and non-conducting particles in a binary granular mixture. The percolation threshold for a number of common regular lattices and for random-packed mono-sized hard spheres are shown in Table 1.

Table 1. Site-percolation threshold and critical-volume fraction for the common three-dimensional lattices and for random-packed hard spheres (Powell, 1979; Scher and Zallen, 1970).

Lattice	Coordination number	Critical volume fraction	Packing density	Critical Percolation fraction
2D				
Triangular	6	0.50	0.9069	0.4534
Square	4	0.59 ± 0.2	0.7854	0.46 ± 0.02
Kagome	4	0.6527	0.6802	0.4440
Honeycomb	3	0.70 ± 0.2	0.6046	0.42 ± 0.01
3D				
diamond	4	0.43 ± 0.03	0.3401	0.146 ± 0.01
sc	6	0.31 ± 0.02	0.5236	0.162 ± 0.01
bcc	8	0.24 ± 0.02	0.6802	0.163 ± 0.01
fcc	12	0.195 ± 0.01	0.7405	0.144 ± 0.01
hcp	12	0.195 ± 0.01	0.7405	0.144 ± 0.01
RPHS	6	0.310	0.59	0.183 ± 0.003

For mono-sized packing, Scher and Zallen (1970) suggested that the volume fraction depends only on the coordination number but not on the lattice structure (Powell, 1979). The dependence of the percolation threshold of a given mono-sized random packing on reciprocal of coordination is given in Fig. 3.

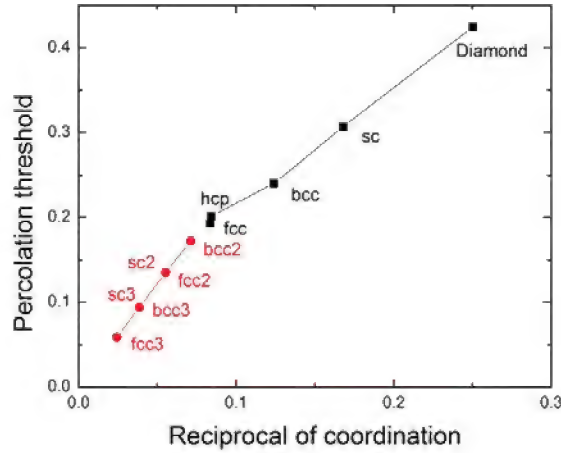


Fig.3. The percolation threshold as a function of the reciprocal of the coordination. The number 2 and 3 after the lattice type indicates coordination to the 2nd and 3rd nearest neighbours (Powell, 1979).

For different-sized binary granular mixtures of randomly packed hard spheres, the determination of the critical volume fraction at percolation threshold can be seen in pioneering work done by Fitzpatrick et al. (1974), Oger et al. (1986), Powell (1979), Suzuki and Oshima (1985), Bouvard and Lange (1991), Kuo and Gupta (1995), and Chen et al. (2009), as is shown in Fig. 4. The percolation threshold was related to one certain coordination number for a range of the size ratios (Kuo and Gupta, 1995). The prediction of the coordination number can be obtained analytically from size ratio and volume fraction. An overview of the different approaches is given in Bertei and Nicoletta (2011).

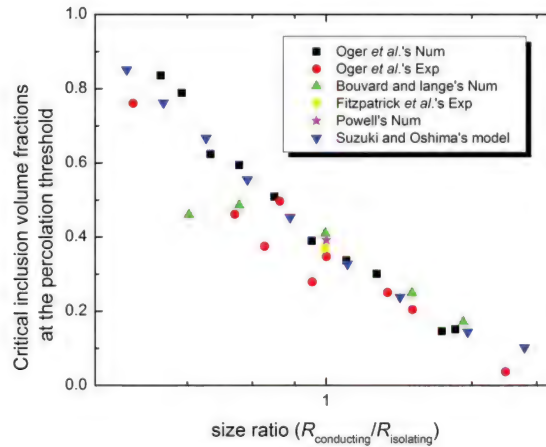


Fig.4. Comparison of experimental and numerical results from literature (Bouvard and Lange, 1991; Fitzpatrick et al., 1974; Martin and Bouvard, 2004; Oger et al., 1986; Powell, 1979; Suzuki and Oshima, 1985) of dependence of critical volume fraction on the size ratio of conducting sphere with respect to the isolating sphere.

The average coordination numbers, Z_0 , of multi-phase randomly packed spheres with size distributions following various rules are found close to 6 (Bernal and Mason, 1960; Suzuki and Oshima, 1985). The value is widely used and is in good agreement with the experimentally determined coordination

number of 6.24 for monosized hard spheres (Pinson et al., 1998). The particle size ratio shows an effect on the coordination number (Bertei and Nicoletta, 2011; Pinson et al., 1998). Mechanical compression can increase the coordination numbers and decrease percolation threshold (Martin and Bouvard, 2004).

2.2.4 Effective conductivity of binary granular mixtures

Percolation probabilities represent the likelihood that particles are clustered in ways that form connected conduction pathways, which is central to the effective functioning of composite structures. For the calculation of the percolation probability of a binary sphere mixture of M phases including conductive phase k and other non-conductive phases, an empirical formula has been put forward (Chen et al., 2009; Costamagna et al., 1998):

$$P = \left(1 - \left(\frac{3.764 - Z_{k,k}}{2} \right)^{2.5} \right)^{0.4}, \quad (2.5)$$

where P is the percolation probability. Here, coordination number, $Z_{k,k}$ (the number of contacts a particle of species k has to particles of the same species), is proportional to the surface-area fraction, δ_k , which is of all k particles and the overall mean coordination number Z_0 . The coordination number $Z_{k,k}$ can be derived by

$$Z_{k,k} = Z_0 \delta_k = Z_0 \frac{\varphi_k / r_k}{\sum_i^M \varphi_i / r_i}, \quad (2.6)$$

where r is the radius of the spheres. The surface-area fraction, δ_k , is defined in terms of the volume fractions φ_k of the k particles:

$$\delta_k = \frac{\varphi_k / r_k}{\sum_i^M \varphi_i / r_i}, \quad (2.7)$$

$$\varphi_k = \frac{n_k r_k^3}{\sum_i^M n_i r_i^3}. \quad (2.8)$$

The determination of effective conductivity through consideration of connectivity within the phase k can be written in the following from:

$$\sigma_{\text{eff}} = K_1 \sigma_0 (P_k - P_T)^\mu, \quad (2.9)$$

where P_k is the volume fraction of the phase k , σ_0 is the conductivity of pure material of phase k , μ is a universal exponent that in three dimensions is 2 (Aharony and Stauffer, 2003), K_1 can be obtained by

$$K_1 = \frac{\gamma}{(1 - P_T)^\mu}. \quad (2.10)$$

So that when $P_k = 1$, $\sigma_{\text{eff}} = \gamma \sigma_0$ where γ is a dimensionless parameter indicating the differences between conductivity of the pure material and the effective conductivity of granular media with identical material (Costamagna et al., 1998).

2.3 Discrete element method

Granular materials are often modelled by either a discrete element method (DEM) or a continuum approach, in order to represent the constitutive behaviour of the materials. Three-dimensional reconstructions of grain assemblies using DEM provide a great deal of quantitative information about the complexity of granular media including the individual inter-particle contact information and the topological features during deformation. DEM is becoming widely accepted as an effective method of addressing engineering problems in granular and discontinuous materials, especially in granular flows, powder technology, and rock mechanics (Cundall and Strack, 1979). However, discrete element methods are relatively computationally intensive, which limits the length of the simulation and the number of particles involved (Radjaï and Dubois, 2011). Due to the vast amount of process parameters and environmental influence, using 3D simulations to decouple the contributions of different variables of microstructural evolution remains challenging (Shearing et al., 2013). The electromechanical coupling has been implemented in DEM approach (Bourbatache et al., 2012; Renouf and Fillot, 2008), suggesting that the electrical computation depends on the mechanical solution. The inter-particle electrical conductance is computed from the contact forces and the resulted in contact area. The overall conductance of the granular assembly can be calculated by solving the group equations obtained by applying Kirchhoff-current law on each individual conductive particle (Creysseles et al., 2009; Ott et al., 2013; Zhai et al., 2017)

2.4 Frequency-dependent network responses

Dielectric relaxation in different materials and systems is one of the most intensely researched topics in physics in recent decades (Jonscher, 1999). The subject of relaxation covers all types of stress relief in solids including dielectric, mechanical, photoconductive, and chemical and so on. It is possible to see a certain commonality of the behaviour for different types of relaxation in the recovery of strain on removal of stress and it implies therefore a time and/or frequency dependence (Jonscher, 1999).

2.4.1 Resistor-capacitor networks

The granular packing can be simplified to a resistor-capacitor (RC) network (Almond et al., 2013; Almond and Bowen, 2004; Almond et al., 2006; Bouamrane and Almond, 2003; Creysseles et al., 2008; Murphy et al., 2006). The network responses have been demonstrated to be robust with respect to microstructural details and component positions. Therefore, all electrical elements including resistor and capacitor are suggested to assign identical values across the network without referring to the contact force distribution for the purpose of simplification (Almond et al., 2006; McCullen et al., 2009).

The frequency dependence of network responses can be divided into three regions. At low frequencies, the AC conductance of the capacitors is much less than that of resistors. The network conductivity is thus dominated by random percolation paths of resistors across the network. At intermediate frequencies, where the conductance of the capacitors and resistors become comparable, all components contribute to the network conductivity. In this emergent frequency range, conductance rises as a power function of frequency. The power law exponent was found to be equal to the capacitive component proportion (Almond and Vainas, 1999; Bouamrane and Almond, 2003; Dyre, 1993; Jonscher, 1977a). At high frequencies, the conductance of the capacitors would far exceed that of the resistors and thus these components can be regarded as short circuits. For cases of high

proportion of capacitors, the network conductance is again determined by the random distribution of all resistors, effectively bound together by shorting capacitors and opening inductors (Clerc et al., 1990).

2.4.2 Mixing rule

The network responses for low and high frequency regions can be analytically estimated based on spectral method (Almond et al., 2013; Jonckheere and Luck, 1998) and the averaging approach (Almond et al., 2013; Milton, 1980; Murphy et al., 2006). A useful approach to view the net admittance for emergent power-law scaling region is known as Lichtenecker's rule (Lichtenecker and Rother, 1931). The scaling properties for simple equivalence rules (Almond and Vainas, 1999) are traditionally given as

$$\sigma_p = \sum_i^N \alpha_i \sigma_i, \quad (2.11)$$

and

$$\sigma_s^{-1} = \sum_i^N \alpha_i (\sigma_i)^{-1}, \quad (2.12)$$

corresponding to parallel and series connections, where σ is the admittance and α the volume fraction. A suitable description of the whole system is a network of connected RC elements, representing each inter-granular contact. The two mentioned simple circuit-equivalence methods of connecting components, represent two opposite extremes. It is also noted that the two equations are of the general form for N -component systems,

$$\sigma^v = \sum_i^N \alpha_i (\sigma_i)^v. \quad (2.13)$$

This suggests a formula that lies somewhere between the limits of equations shown above, with v of $+1$ and -1 for series and parallel connections, respectively. Considering the logarithmic mixing rule (Almond et al., 2006; Murphy et al., 2006), we can further simplify the conductivity of a system with two distinct types of components, such as multi-phase materials or an RC circuit, to

$$\log(\sigma) = \alpha_1 \log(\sigma_1) + \alpha_2 \log(\sigma_2), \quad (2.14)$$

$$\sigma = \sigma_1^{\alpha_1} \sigma_2^{\alpha_2}, \quad (2.15)$$

where $\alpha_1 + \alpha_2 = 1$. For an RC circuit network, the individual admittances are $\sigma_1 = 1/R$ and $\sigma_2 = i\omega C$, corresponding to resistors and capacitors. The net admittance may then be expressed as

$$Y = \left(\frac{1}{R}\right)^{\alpha_1} (i\omega C)^{\alpha_2}, \quad (2.16)$$

where $(i\omega C)^{\alpha_2}$ can be further expressed by $(\omega C)^{\alpha_2} \left[\cos\left(\frac{\alpha_2 \pi}{2}\right) + i \sin\left(\frac{\alpha_2 \pi}{2}\right) \right]$. The real part of the admittance is the conductance,

$$G = \text{Re}(Y). \quad (2.17)$$

Taking the logarithm of both sides leads to

$$\log(G) = \alpha_2 \log(\omega) + B, \quad (2.18)$$

where $B = \left[-\alpha_1 \log(R) + \alpha_2 \log(C) + \log\left(\cos\left(\frac{\alpha_2 \pi}{2}\right)\right) \right]$. This suggests a power law relationship between conductance and impedance, with respect to frequency for networks of randomly positioned R and C components. The parameter, α_2 , describing the proportions of capacitive components, determines the exponents of the power functions.

2.5 Contact mechanics

A wide range of relaxation phenomena are strongly associated with interfacial processes, such as in metal–insulator, semiconductor–insulator, electrode–electrolyte and similar systems. Specifically, the interfacial roughness is fundamentally important in the conductance in studied granular systems, by determining the inter-particle conductance that act as the RC network element.

A variety of electrical contact resistance models have been developed since rough surface contacts were first considered by Archard (1957), and further developed by Ciavarella et al. (2004), Kogut and Komvopoulos (2004), Jackson and Streater (2006), and Jackson et al. (2015). In experiments, many mechanisms of surface structure evolution have been observed during electrical conduction through rough interfaces, including dielectric breakdown of oxide layers, localised current-induced welding, chemical disorder arising with random composition and oxidation processes in corrosive environments (Creysse et al., 2007; Falcon et al., 2004). The complex multi-scale morphologies exhibited by rough surface structures give rise to difficulties in the direct quantitative evaluation of real interfacial contact area, and therefore the contact conductance. The interfacial properties and contact mechanics employed in existing models of granular systems are typically simplified (Béquin and Tournat, 2010; Bourbatache et al., 2013; Bourbatache et al., 2012).

A single electrical contact between two grains can be represented by a resistor, capacitor or inductor, depending on the inter-particle distance and force. Separating conductive spheres can be simply considered as pairs of capacitors. For widely used metallic materials (Bourbatache et al., 2012; Radjaï and Dubois, 2011; Renouf and Fillot, 2008), a thin surface oxide layer acting as a resistive film with typical thickness ranging from 1 to 100 nm (Kikuti et al., 2004; Proust et al., 2015) can be usually found. Consequently, these dielectric layers cause contacts to be non-conductive, under conditions of low loads, effectively acting as capacitors. When two spheres are compressed with sufficient pressure, surface asperities penetrate the oxide layer thus forming small metal-to-metal contact patches. This leads to the conduction of electrical current with a high-level resistance, governed by the constriction resistance (Greenwood, 1966; Holm, 1967; Mikrajuddin et al., 1999). The contacting interfaces between grains tend to exhibit complex geometries and structures, over a wide range of length scales, governing physical properties and interfacial phenomena and giving rise to constriction resistance. The constriction resistance due to the convergence and divergence of current flow at a single contact, results in a high resistance state. When the radii of contacting asperities are comparable or smaller than the average electron mean free path, electrons can only travel ballistically across the micro-contacts, e.g., tunneling (Fisher and Giaever, 1961; Mikrajuddin et al., 1999). Consequently, tunneling through the oxide layer contributes to the conduction at this stage (Kogut and Komvopoulos, 2004).

Under increasing compression, the numerous independent contacting spots increase in size merging into larger contacts. When the effective radius of a contacting asperity becomes larger than the mean free path of electrons, Holm contact (Jackson et al., 2012) will be the dominant transport mechanism, resulting in a relative lower resistance. With further increasing inter-particle force, large zones of metal-to-metal contact appear, likely establishing metal chains along several contacting grains, presenting inductive properties. Noticeably, the inductive phase may not appear for metals with thick oxide layers, such as RuO_2 , even with high levels of compression (Grimaldi et al., 2002), and for fine powders (Creysseles et al., 2008) with high surface-to-volume ratios and hence a high proportion of oxide, the inductive phase may not be observed, under insufficient pressure.

3 Contact Mechanics at Rough Interfaces

In a range of applications employing granular materials, such as geothermal energy (Lund et al., 2005), soil probing (Deschamps et al., 2000), sustainable agriculture (Lal, 2009), slope stability (Gunzburger et al., 2005), and foundations for high-rise buildings (Jinli, 2006), interfacial characteristics at the finest length scales strongly impact overall system performance. Normal contact stiffness is one of the most important physical quantities related to the generalized force displacement behaviour of rough surfaces in contact. The contact stiffness indicating the evolution of true contact area depends on the applied pressure and is of notable importance for the study of systems involving the physical interactions of multiple bodies including granular matter, electrode contacts, and thermal contacts, where the interface-localized structures govern overall system performance. In this chapter, we experimentally and numerically investigate the influence of surface topology on normal contact stiffness.

3.1 Paper 1: Experimental investigation on contact stiffness

This first paper in this chapter shows an experimental investigation into the effects of roughness and fractality on the normal contact stiffness of rough surfaces. We considered different types of rough surfaces altered by polishing and by five surface mechanical treatments using different sized particles. Surface structures were characterised by conventional first- and second-order roughness parameters and fractal dimension calculated based on multiple methods. We employ the nanoindentation to evaluate the normal contact stiffness with flat tips. The results of our original experiments address existing controversy in the literature and show that contact stiffness at rough interfaces follows a power-law function with respect to normal force over a wide range of applied loads. We have further connected the experimentally obtained power-law exponent and amplitude to surface structure characteristics. These correlations are useful in establishing and validating predictive models for contact problems for a wide range of rough surfaces, including the curved rough contacts. In Chapter 5 and 6, the obtained correlations between surface structure and interfacial stiffness will be integrated into granular packings, which can be regarded as multi-contact systems, to understand stress-dependent responses of granular materials.

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The Role of Surface Structure in Normal Contact Stiffness

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Abstract The effects of roughness and fractality on the normal contact stiffness of rough surfaces were investigated by considering samples of isotropically roughened aluminium. Surface features of samples were altered by polishing and by five surface mechanical treatments using different sized particles. Surface topology was characterised by interferometry-based profilometry and electron microscopy. Subsequently, the normal contact stiffness was evaluated through flat-tipped diamond nanoindentation tests employing the partial unloading method to isolate elastic deformation. Three indenter tips of various sizes were utilised in order to gain results across a wide range of stress levels. We focus on establishing relationships between interfacial stiffness and roughness descriptors, combined with the effects of the fractal dimension of surfaces over various length scales. The experimental results show that the observed contact stiffness is a power-law function of the normal force with the exponent of this relationship closely correlated to surfaces' values of fractal dimension, yielding corresponding correlation coefficients above 90 %. A relatively weak correlation coefficient of 60 % was found between the exponent and surfaces' RMS roughness values. The RMS roughness mainly contributes to the magnitude of the contact stiffness, when surfaces have similar fractal structures at a given loading, with a correlation coefficient of ~95 %. These findings from this work can be served as the

experimental basis for modelling contact stiffness on various rough surfaces.

Keywords Contact mechanics · Contact stiffness · Rough surfaces · Fractal dimension · Nanoindentation

Introduction

In contact mechanics, surface morphology plays an essential role in determining how solids interact with one another, with significance in many processes including friction, wear, thermal and electrical conduction [1–3]. The classic Hertzian contact theory considers elastic solids with smooth profiles of curved surfaces connecting the applied force and indentation depth, by assuming a distribution of normal pressure in the contact area. However, this assumption neglects topological features of natural surfaces and the interaction between individual contact regions [2]. The existence of surface roughness results in only a certain number of peaks or asperities being in contact. Since the true contact area at an individual asperity can be of nanoscale dimensions, the real contact area between two surfaces is typically orders of magnitude smaller than the apparent or nominal contact area. As early as the 1950s, Bowden and Tabor had already recognized the significance of the surface roughness of contacting bodies in contact mechanics. Since then, the mechanics of rough interfaces have been broadly studied on the basis of early contributions by Archard [4] and Greenwood and Williamson [5]. In the past few years, many studies have been carried out to interpret parameters of contact area and normal contact stiffness under various loading forces, for different surface characteristics [6–9]. It has been theoretically and numerically found that in non-adhesive contact, the contact area between rough elastic surfaces depends linearly on the normal force and is inversely

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proportional to elastic modulus and the root mean square (RMS) slope of the surface [10–12].

The true interfacial contact area along with roughness parameters could, at some level, be utilised to interpret the mechanics of rough surfaces subjected to an applied normal load [13, 14]. However, the determination of the real contact area between contacting bodies through experimental measurement is challenging because most natural surfaces exhibit features across a wide range of length scales [15, 16]. Rough surfaces possess complex morphologies and geometries that are difficult to quantify definitively.

Numerical analyses, using molecular dynamics, boundary element methods, etc., have been conducted and generally support the proportionality between normal force and contact stiffness, as was first mentioned in the Greenwood-Williamson model [5, 8, 10, 17]. However, other studies have reported that, for small to medium loads, the logarithm of stiffness exhibits close proportionality to the logarithm of the applied normal force, i.e. the contact stiffness k is a power function of the normal force F_N : $k \propto F_N^\alpha$, which differs from the prediction of Greenwood-Williamson model and the work mentioned above. This issue has been the subject of controversy and discussion in the field of contact mechanics for some years [7, 18–21].

Several related experiments have been conducted on multiscale rough surfaces to gain information pertaining to interfacial behaviour. Buzio et al. [22] employed atomic force microscopy (AFM) to explore the role of surface morphology in contact mechanics at the nanoscale. This approach was selected owing to its high sensitivity with respect to vertical displacements and normal loads. However, observations of morphological effects with nanometric tips (for example, sharp AFM tips) are encumbered by technical difficulties, e.g., uncontrolled long-range adhesive forces dominate incipient contact while adsorbed water and contaminants may smooth atomic corrugation [23, 24]. The digital image correlation and ultrasound techniques have been employed to measure the contact stiffness of real engineering interfaces, revealing that contact stiffness depends upon three main factors: contact pressure, surface roughness, and surface hardness [25]. Relationships between hierarchical surface structures, loading conditions and contact mechanics have been studied experimentally and computationally using a variety of other macroscopic approaches and a range of materials [21, 26–32]. However, significant difficulties remain in relating parameters of contact stiffness to surface structure descriptors.

The surface morphology of materials is known to be of fundamental importance in governing physical properties and interfacial phenomena. Consequently, there exists an ever increasing number of experimental techniques for the two and three dimensional characterisation of rough surfaces along with a broad range of numerical descriptors to quantify these structures using parameters such as roughness, skewness,

kurtosis, curvature, slope, and fractal dimension. Among these surface descriptors, the RMS roughness, RMS slope and fractal dimension are of considerable value in comparative analysis and quantitative characterisation of surface structures. The RMS slope is commonly chosen as a higher order surface descriptor, which can be used to interpret surface phenomena. For example, optical and tribological properties at rough interfaces have been varyingly correlated with RMS slope [33, 34]. Moreover, the fractal dimension of surfaces is considered to be an important descriptor useful in representing realistic three-dimensional surface features over a wide range of length scales [35, 36]. The fractal dimension, a cross-scale surface descriptor that incorporates localised and macroscopic morphological surface behaviour, has been attracting increasing attention in the characterisation of surfaces and particles across a wide range of fields including stereology, powder technology, geology, metallurgy, and computer vision [37, 38]. The advantage of using surface fractality as a cross-scale descriptor stems partly from the tendency of first-order descriptors (e.g. RMS roughness) to be dominated by the highest scale features, while secondary descriptors (e.g. RMS slope) tend to be dominated by the finest scale surface characteristics.

In spite of the significant progress that has been made in nanoindentation and surface morphology characterisation, which facilitate the mechanical and morphological analysis of surfaces at micrometre and nanometre scales, the existing experimental results of the structure-dependence of contact behaviour of surfaces with random multiscale features are limited. In this work, we employed flat tip nanoindentation to directly observe normal contact mechanics at rough surfaces and, in conjunction with 3D profilometry, established relationships between hierarchical surface structures and exhibited contact stiffness.

Method

Sample Preparation and Characterisation

In this paper, we considered round disk samples made of aluminium alloy 5005 owing to this material's suitable chemical stability and deformability. Three surface treatment methods were applied: (1) polishing, (2) abrasive blasting and (3) surface mechanical attrition treatment (SMAT). Because the aluminium alloy used is relatively soft and ductile, the surfaces of these samples can be efficiently modified through standard polishing treatment, bead blasting and SMAT procedures. Samples with polished surfaces were prepared using sequential grinding and polishing steps with final polishing using 1 μm diamond suspension. The other two methods employed here accomplish physical modification of the surface details at different length scales depending on the size of the particles

used in the treatment. Both treatments utilised the impact of high-speed particles on specimen surfaces. Specifically, the operation of bead blasting is propelling forcibly a stream of fine glass beads to impact and modify the surface morphology. The average sizes of the two selected groups of glass beads used in the blasting treatment of the polished surfaces were 50 and 300 μm , respectively. SMAT alters surface features using an excitation mechanism to accelerate smooth steel balls and project them on the prepared sample surfaces. The surface treatments employed will introduce changes in the surface-localized microstructure, and thus may alter the material hardness. However, the material elasticity will not be strongly affected by the surface treatments, e.g. SMAT [39–42]. In our work we used three different SMAT processes using 1, 2 and 3 mm diameter balls, respectively. Figure 1 compares the samples achieved through the different methods, using a scanning electron microscope (SEM, Zeiss ULTRA plus), at the identical magnification. It can be seen clearly that samples after blasting treatment using glass beads of 50 μm present more complex and rougher texture.

To further quantitatively analyse the surface morphology, the treated aluminium surfaces were scanned using an optical surface profilometer (NanoMap 1000WLI). A standard 1024×1024 topographical imaging procedure with a vertical scanning range of $20 \mu\text{m}$ was applied with a green LED light source (with a wavelength of $\sim 500 \text{ nm}$) to obtain three-dimensional topographical maps of the specimen surfaces. Subsequently, surface roughness parameters were determined from the digitised surface data across multiple scans of $1 \times 1 \text{ mm}^2$ for each individual sample. The sample surfaces were primarily characterised through two roughness descriptors, RMS roughness and RMS slope, which are two widely used surface descriptors for the characterisation of rough interfaces.

For the purpose of quantifying and comparing the prepared surfaces across a wide range of length scales, a grid of unit dimension L is superimposed in order to mesh the obtained digitised surface into a number of triangles. The variance of surface areas of different samples was calculated at different digital resolutions of the scans, as shown in Fig. 2. For example, when $L=X/4$ with X being the total scan length, the surface is covered by 32 triangles of different areas inclined at various angles with respect to the projected plane. The areas of

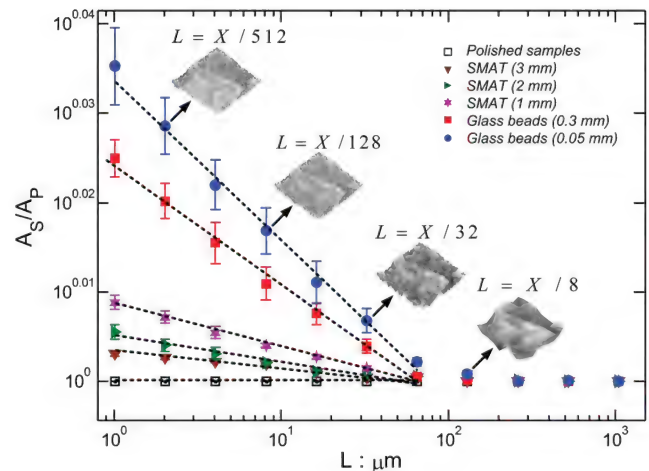
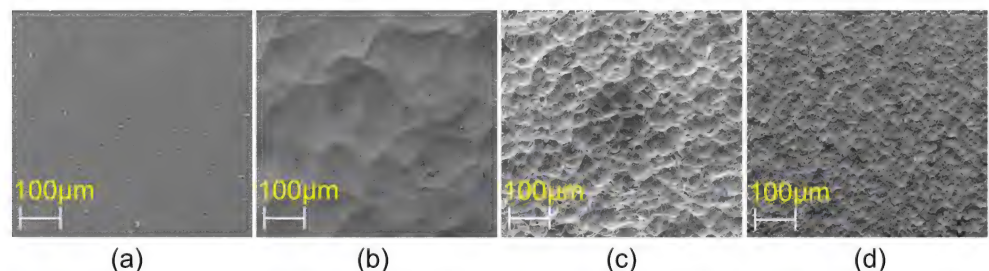


Fig. 2 The calculated sample surface areas at different scales. The insets show the digitised scans used to calculate the surface area for the samples that underwent sand blasting using 50 μm -sized glass beads

all triangles are calculated and summed to obtain a total surface area A_s for a given value of L . These triangular elements have an equal projected triangle area, although their real areas are different and the total surface area A_s is larger than the projected area of the scanned sample A_p . The grid size is then decreased by a successive factor of 2, and the previously mentioned process continues until L corresponds to the distance between two adjacent pixel points, i.e., the highest resolution of the digitised data, which is 1 μm for the optical profilometer employed in the present work. For all of the six sample surfaces, the obtained surface areas A_s at various L values exhibit an increasing trend as the grid is made finer with a smaller value of L . Of the six types of surfaces, the surface area of the sample blasted by glass beads shows more evident variation at different scales than that of the SMAT prepared samples or polished specimen. A power law relationship is found between the calculated surface area and the length resolution of digitised scan, in the range from 1 to 1000 μm . In other words, the sample surface structures exhibit self-affinity over a certain range of length scales, which can be characterised by the surfaces' fractal dimension.

In this paper, the concept of fractal dimension provides a useful method for representing rough surfaces in terms of cross-scales analysis. A number of existing algorithms have

Fig. 1 SEM images of aluminium samples with different surface treatments: (a) polishing treatment; (b) SMAT using 2 mm-sized steel balls; (c) blasted with 300 μm -sized glass beads; (d) blasted with 50 μm -sized glass beads



been fully developed to determine surface fractal dimension from digitised surface data [38, 43]. Here, we used two methods to obtain the fractal dimension: (1) Scaled triangulation; (2) Cube counting. The process of the triangulation method based on cross-scale comparison is illustrated in Fig. 2, demonstrating the calculated surface areas at various scales. The error bars are achieved over five samples for each type of surface. The box-counting approach is also applied here as a comparison [44, 45]. For both methods, an anisotropic scaling is applied to normalise three-dimensional scans, making both methods suitable to self-affine surfaces.

As shown in Table 1, six typical surface types with distinct surface details were prepared and characterised prior to nanoindentation tests. For each surface type, the mean values of roughness descriptors were obtained over ten scans and the corresponding standard deviations showed these surface measures were representative. The resulting values are reported as indicated in Table 1 including the mean RMS roughness, RMS slope, and fractal dimension. The two calculation methods used here to obtain the values of fractal dimension are principally in agreement with each other, with the values of fractal dimension inverse to the sizes of particles applied in SMAT and glass-bead blasting treatments. Moreover, the RMS slope and fractal dimension obtained from both methods show similar trends, i.e., smaller particles used to modify the surfaces lead to larger RMS slope and fractal dimension values. As for the RMS roughness, results show that the parameter has no clear correlation with varying particle sizes.

Contact Stiffness Measurement Using Nanoindentation

Surface contact stiffness of aluminium samples with different surface morphology was analysed by means of nanoindentation tests with three flat indenter tips with different diameter sizes: 54.1, 108.7, and 502.6 μm (SYNTON-MDP, FLT-D050, FLT-D100, and FLT-D500), as shown in Fig. 3. The tips have sizes comparable with the size of the particles used to modify the surface of the specimens and the roll-off wavelength found in the power spectra of the surfaces ranging from 50 to 250 μm [43]. The three flat indenter tips were also

examined by profilometry, and were found to exhibit an average RMS roughness of less than 0.04 μm , and thus in the following calculations, the tip surfaces were assumed to be ideally smooth. The reason for choosing flat tips is that the apparent contact area under the tip does not change with respect to the indentation depth, which is not the case for spherical or Berkovich tips. All the tips and specimens under tests were properly cleaned using water and compressed air to remove any embedded grains and particles. Cleaning with ethanol and heat treatment (around 120 $^{\circ}\text{C}$) were also applied to remove surface contamination and physisorbed moisture. When the flat indenter tip first contacts the sample surface, the actual contact area is only a small fraction of the nominal contact area. The asperities of the sample surface at contact regions are squeezed against the flat tip, deforming elastically or plastically. For the purpose of evaluating only the elastic responses, 10 partial unloading procedures were employed by decreasing the applied load by 10 %, in the course of each individual test. After each unloading stage, the loading level rose by a factor of two to the next unloading stage, until the load reached 500 mN where the last unloading was performed. All the values of stiffness were determined by averaging over 10 indentation tests at different positions on each surface type. Analogous analytical procedures have been implemented experimentally at aluminium interfaces in the past using a method of ultrasonic loading [46] and computationally in the framework of finite element analyses [47]. The unloading stiffness k (in units of N/m) was defined as the slope of the unloading curve, $k = dP/dS$, where P designates the load and S is the indentation depth, as indicated in Fig. 3. Subsequently, the reduced elastic modulus E_r can be derived from the measured unloading stiffness through the relation:

$$E_r = \frac{\sqrt{\pi}}{2} \frac{k}{\sqrt{A}}, \quad (1)$$

where A is the nominal contact area of the indenter tip. Equation (1) is a fundamental equation for assessing the elastic properties in nanoindentation tests, and has been shown to be equally applicable in cases of elastic–plastic contact [48–51]. The applicability of this formula when plastic deformation

Table 1 Sample surface characterisation with different treatments

Surface treatment	RMS Roughness $R_{RMS}/\mu\text{m}$	RMS slope R_s	Fractal dimension D_{f1} / Triangulation	Fractal dimension D_{f2} / Cube counting
1. Polish	0.05712 $\pm$ 0.005431	0.009101 $\pm$ 0.0009640	2.093 $\pm$ 0.06176	2.024 $\pm$ 0.04101
2. SMAT3mm Particle size: 3 mm	4.012 $\pm$ 0.3674	0.08280 $\pm$ 0.007314	2.185 $\pm$ 0.0351	2.149 $\pm$ 0.02543
3. SMAT2mm Particle size: 2 mm	2.730 $\pm$ 0.2554	0.07633 $\pm$ 0.00397	2.228 $\pm$ 0.01987	2.156 $\pm$ 0.01308
4. SMAT1mm Particle size: 1 mm	1.152 $\pm$ 0.3124	0.04864 $\pm$ 0.01137	2.337 $\pm$ 0.01818	2.233 $\pm$ 0.01659
5. GB300 μm Blasted by glass beads of 300 μm	4.179 $\pm$ 0.1943	0.2244 $\pm$ 0.01537	2.551 $\pm$ 0.02170	2.424 $\pm$ 0.02572
6. GB50 μm Blasted by glass beads of 50 μm	2.970 $\pm$ 0.2759	0.2023 $\pm$ 0.01022	2.626 $\pm$ 0.01736	2.351 $\pm$ 0.03633

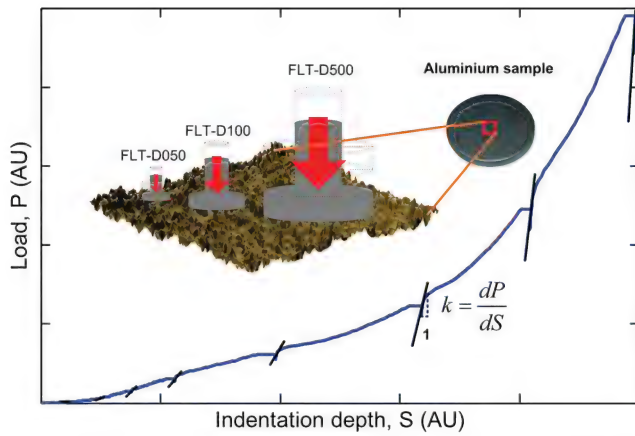


Fig. 3 Typical loading-displacement curve of nanoindentation tests on the sample surfaces, in arbitrary units (AU). Partial unloading processes were applied to isolate elastic contributions to contact stiffness under different loading levels. Three flat indenter tips used in the experiments (FLT-D050, FLT-D100, FLT-D500) are also illustrated for comparison

occurs during the indentation experiments has been the subject of numerous studies [52, 53].

Considering the fact that the measured reduced elastic modulus E_r includes contributions from both the tested specimen and the indenter tip, the contact stiffness, E_c (in the unit of N/m^2), is estimated from

$$\frac{1}{E_r} = \frac{1-\nu_c^2}{E_c} + \frac{1-\nu_i^2}{E_i}, \quad (2)$$

where the subscript i indicates the property of the indenter tip material, the subscript c the properties of the tested specimen and ν is the material's Poisson's ratio. For the diamond indenter tips used in this research, E_i and ν_i are typically 1140 GPa and 0.07, respectively. For the aluminium samples employed here, ν_c is set as 0.3.

Results and Discussion

Experimental Data

Figure 4 presents the typical measured contact stiffness of rough surfaces created by sand blasting treatment with 50 μm diameter glass beads (Sample GB50 μm) obtained from nanoindentation tests with three tips of different sizes. The stress was calculated from the ratio of loading force to the projected area of the indenter tips. Figure 4 shows that, by using different sized tips, the stress range extends over several orders of magnitude. With the same maximum force (~ 500 mN) provided by the nanoindenter, the maximum stress produced with FLT-D050 was around 100 times larger than that of the FLT-D500. The stress provided by all the three indenter tips ranged from 0.005 to 214.6 MPa, crossing five orders of magnitude with the FLT-D500, FLT-D100, FLT-

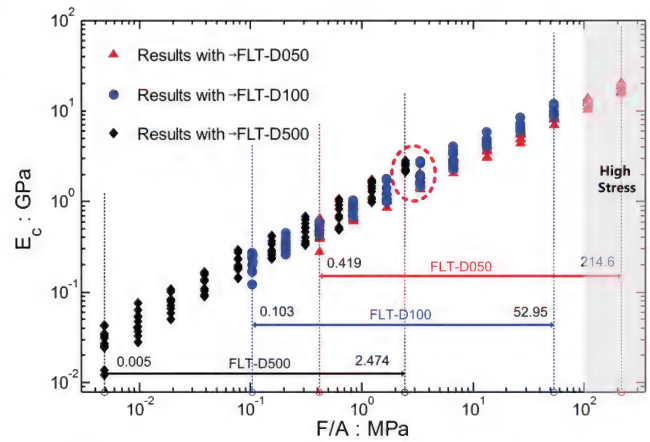


Fig. 4 Contact stiffness, E_c , measured from rough surfaces with sand blasting treatment of glass beads of 50 μm diameter (GB50 μm). Raw data are from multiple indentation tests at different loading levels, F/A , where F is the normal force and A denotes the projected area of the flat tip

D050 corresponding to the ranges (shown in Fig. 4) of 0.05 to 2.474 MPa, 0.103 to 52.95 MPa, and 0.419 to 214.6 MPa, respectively. The contact stiffness measured over this range of applied stresses varies approximately from 0.01 to 55 GPa.

It can be seen clearly that the measured contact stiffness increases with the loading force, for all tested samples, as shown in Fig. 5. Although, as mentioned in the introduction to the present work, it is the subject of some controversy, the contact stiffness has been predicted by certain reports to exhibit a power-law relation with the applied normal force, which can be described as $E_c \propto F_N^\alpha$, where F_N is the normal force applied on the surface and α is the exponent of the power function [13, 19, 21, 22]. At the same applied stress level, the surfaces after SMAT (Sample 2–4) or sand blasting treatment (Sample 5–6) show a smaller value of contact stiffness with respect to that of the polished surface (Sample 1). The surface blasted with glass beads of 50 μm diameter (Sample 6)

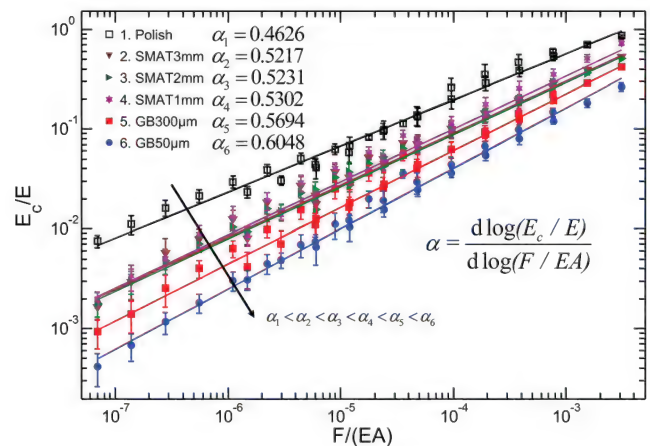


Fig. 5 Curve fitting for the normalised stiffness, E_c/E , and normalised applied load, $F/(EA)$, for six tested surfaces, with E being the Young's modulus of the tested material, and A the apparent contact area

presents the lowest contact stiffness of all the six types of surfaces. It is also found that the surfaces with the highest values of RMS slope and fractal dimension exhibit comparatively lower contact stiffness in the range of applied stress levels.

As the loading force increases towards the high stress regimes, the slope of the measured contact stiffness decreases. The trend is more pronounced as the normal stress approaches the bulk compressive strength of the aluminium alloy (shown in Fig. 4). This trend matches the expectation that with deformation of individual asperities the contact stiffness will converge to a stable value, close to the elastic stiffness of the bulk material.

For each surface, the measured contact stiffness obtained through one indenter tip briefly coincides with that of the other two indenter tips of different sizes in the overlapping stress ranges. However, compared to the results with smaller tips, the measured contact stiffness obtained with larger indenter tip (FLT-D500) tends to be larger at similar stress levels. A typical example is shown in the red dashed circle in Fig. 4. The measured contact stiffness at the maximum stress for FLT-D500 is evidently larger than the result achieved with FLT-D050 under a similar stress level. The indentation of a single asperity or fewer asperities, the likely result of the use of the 50 μm tip, occurs with lower apparent stiffness owing to the presence of unconstrained neighbouring surface features. While the exact nature of deformation mechanisms differs between materials it is likely that this phenomenon occurs to varying extents across a broad range of systems.

An alternative explanation to the increased stiffness observed with the use of larger indenter sizes is the adhesion forces between the tip and the rough surface, which are strongly affected by the true contact area. A larger tip tends to yield a relatively higher level of adhesion, resulting in higher bond strength and larger apparent contact stiffness. The adhesive force can be observed even in the absence of capillary bridges if the surfaces are sufficiently smooth [2, 54]. An estimation of bond strength can be made at the instant of surface separation at the end of the indentation tests, found typically within the range of 10 MPa [55, 56]. In our situation, an adhesive interaction between the indenter tip and the testing specimen could not be eliminated even after proper cleaning and drying processes, and it can potentially impact the measurements, particularly for cases of polished samples subjected to high applied loading forces.

Another factor that is likely to influence the measured contact stiffness in nanoindentation experiments is the oxide layer on the surfaces of aluminium samples. Aluminium alloys ubiquitously exhibit a thin passivated oxide layer arising from reaction with atmospheric oxygen. This nanoscale oxide layer exhibits locally divergent mechanical properties in a region of thickness typically less than 10 nm [57, 58]. In this research, the penetration depth of the nanoindentation test ranges from

1000 to 5000 nm. The smallest depth is observed in one test accomplished with FLT-D500 on a polished sample, where 1025.6 nm is achieved with 500 mN being the maximum force. The influence of the oxide layer is thus expected to be of limited significance owing to the considerable difference in the thickness of the oxide layer and indentation penetration depth.

To compare the contact stiffness of different surfaces, we converted the obtained values for contact elastic modulus E_c through equations (1) and (2) to non-dimensional values E_c/E by dividing the value of the Young's Modulus of aluminium alloy 5005, $E=69.5$ GPa. The non-dimensional stress is defined as $F/(EA)$, where A is the projected area of the corresponding tip. Note that the fitting curves are achieved, excluding the contributions from the measured stiffness under stress levels higher than 100 MPa, where the surface shows an apparent yield phenomenon. The slope α of the fitting curve can be defined as the exponent of the power function between the normalised contact modulus and normalised stress. For all the six sample surfaces, the value of the exponent α varies from 0.4626 to 0.6048, changing as the fractal dimension and the RMS slope increase. As a comparison, this typical value in cases of Hertzian contact of two elastic spheres is 1/3. The power-law relationship found here experimentally is in good agreement with previous theoretical predictions on a quantitative basis [13, 18, 19, 21].

Correlation Analysis

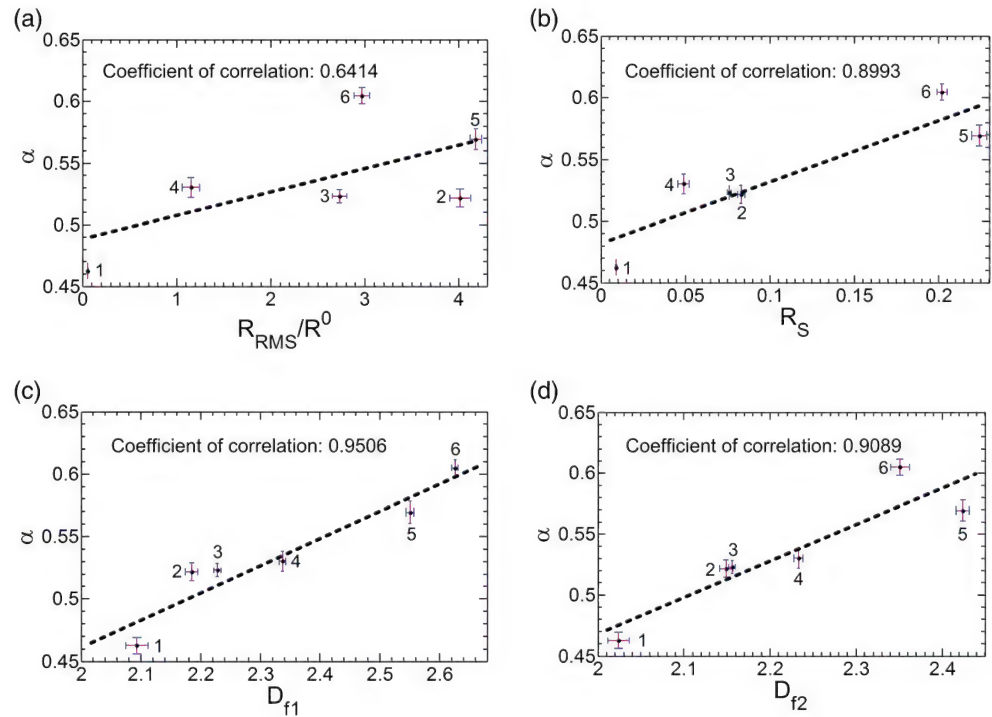
From the normalised data shown in Fig. 5 the relation between the contact modulus E_c and applied loading force F can be described as

$$\frac{E_c}{E} = \beta \left(\frac{F}{EA} \right)^\alpha \quad (3)$$

The exponent α and amplitude β are assumed to be constant for a given surface structure. Using this form, we can obtain a set of coefficients, α and β , corresponding to the tested surfaces from the measurements in Fig. 5.

Correlations between the exponent α and the evaluated surface parameters are described in Fig. 6, where, for all tested surfaces, the exponent α is plotted against the aforementioned surface descriptors, i.e., (1) RMS roughness; (2) RMS slope; (3) fractal dimension found from a triangulation method, and (4) fractal dimension found using a box counting method. Specifically, the exponent α achieved across a wide range of stress values illustrates a relatively weak correlation to values of RMS surface roughness. While in contrast, this exponent correlates more closely to values of the fractal dimension with the corresponding correlation coefficients above 90 %. In Fig. 6(b), a correlation coefficient of around 90 % has also been found with the RMS slope, but the exponent α of the

Fig. 6 Correlations between the stiffness variation exponent α and roughness parameters: (a) Non-dimensionalised RMS roughness R_{RMS}/R^0 , with $R^0 = 1 \mu\text{m}$; (b) RMS slope; (c) Fractal dimension obtained with the scaled triangulation method; (d) Fractal dimension obtained with the cube counting method. Horizontal and vertical error bars show the corresponding standard deviations from the surface measurement and curve fitting. The sample number shown in the figures can be referred to Table 1



SMAT samples yields similar values and does not show strong correlation to the RMS slope. This, to a certain degree, is supported by numerical investigations of these descriptors reported in the literature, which, it should be noted, focused mainly on the evolution of contact area and surface separation rather than stiffness [59].

Figure 7 illustrates the significance of RMS roughness, RMS slope and fractal dimension in governing the coefficient β for the contact stiffness measured with flat tipped diamond indenters. It is found that the RMS roughness dominates β with the coefficient of correlation being -95% , whilst the RMS slope and the fractal dimension are not as well correlated

Fig. 7 Correlations between the coefficient β and roughness parameters: (a) Non-dimensionalised RMS roughness R_{RMS}/R^0 , with R^0 being $1 \mu\text{m}$; (b) RMS slope; (c) Fractal dimension obtained with the scaled triangulation method; (d) Fractal dimension obtained with the cube counting method. Horizontal and vertical error bars show the corresponding standard deviations from the surface measurement and curve fitting. The sample number shown in the figures can be referred to Table 1

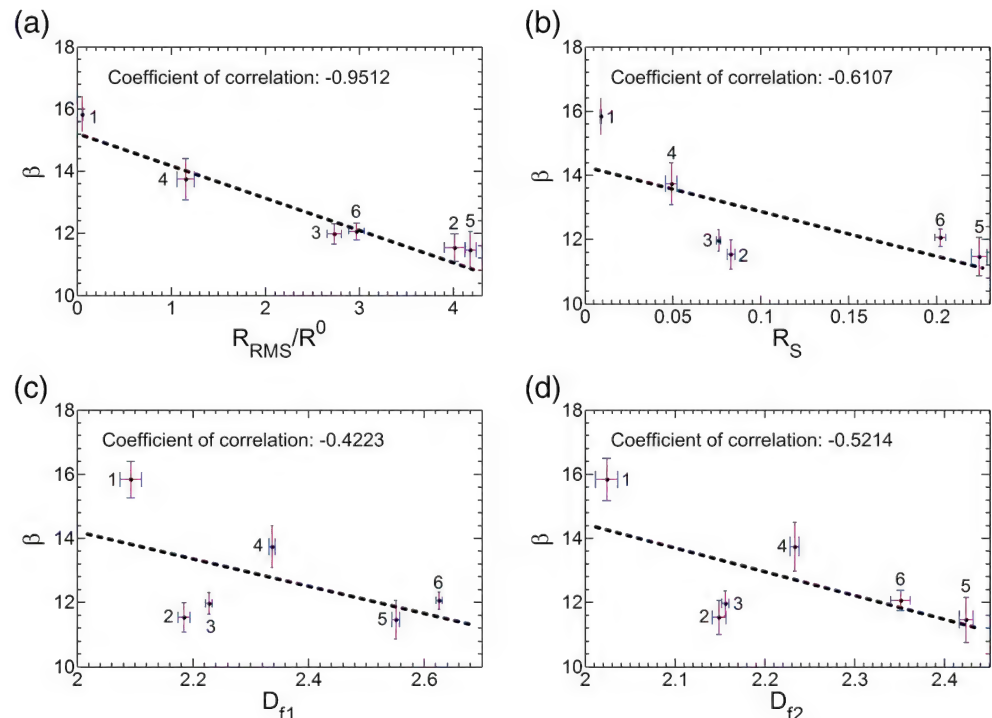


Table 2 Correlations between the stiffness parameters and surface roughness parameters

	RMS Roughness $R^*=R_{RMS}/1\ \mu\text{m}$	RMS slope R_S	Fractal dimension D_{f1} Triangulation	Fractal dimension D_{f2} Cube counting
α	$\alpha=0.019R^*+0.487$	$\alpha=0.503R_S+0.481$	$\alpha=0.217D_{f1}+0.029$	$\alpha=0.301D_{f2}+0.133$
β	$\beta=-1.02R^*+15.3$	$\beta=-14.1R_S+14.27$	$\beta=-4.28D_{f1}+22.8$	$\beta=-7.61D_{f2}+29.7$

with this coefficient. The RMS roughness is a parameter indicating primarily the vertical scale of the rough surface. If all the tested surface structures, along with indentation depths, were to be stretched or compressed by a selected factor yielding identical roughness ranges and the same value of β , the contact behaviour would be mainly controlled by the fractality of the surface as described by the value of its fractal dimension. The fitting functions for the stiffness parameters, i.e., α and β , using surface roughness parameters, including RMS roughness, RMS slope and fractal dimension are detailed in Table 2.

Similar formulas have been used as the theoretical approximation for the contact stiffness from Pohrt and Popov [19], Jiang et al. [21] and Komvopoulos and Ye [7]. In all the aforementioned theories, the relationship between the stiffness and pressure was found to follow a power law, with the exponent strongly correlated to the value of fractal dimension. Pohrt and Popov [19] established a power law based on boundary element simulations for a rigid square indenter over a range of normal loads spanning four orders of magnitude, concluding that the contact stiffness for low to medium loading conditions is most appropriately approximated by a power-law dependence with the exponent ranging from 0.51 to 0.77, corresponding to a variation of fractal dimension from 2 to 3 [19]. It is worth noting that the approximated value of the exponent α is $0.2567D_f$, which is rather close to the value of $0.2168D_{f1}$ found experimentally in the current work. Additionally, Jiang et al. [21] have presented a fractal model which can be applied to analyse the contact stiffness of machined plane joint surfaces, in which the exponent α of the power law is described as $D_f/(3-D_f)$. Under a given load, smaller values of both surface roughness and fractal dimension can increase the true contact area, thereby resulting in a larger number of micro-contacts yielding elastic deformation, which, in turn, increases the normal and tangential contact stiffness.

Conclusions

In this work, the effects of surface roughness and fractality on the normal contact stiffness were experimentally demonstrated for various rough surfaces. A wide range of applied stress across five orders of magnitude is achieved using three flat indenter tips of various sizes. The results in our experiments show that the contact stiffness follows a power-law function with respect to the normal force, with the exponent of this

relationship ranging from 0.4626 to 0.6048, corresponding to the polished surface and the surface treated with the finest beads. This relationship is then described through an expression with two parameters, simplified as $E_c/E=\beta[F/(EA)]^\alpha$. Through correlation analyses, we connected the experimentally obtained coefficients, i.e., the exponent α and amplitude β , to the characterisation of rough surfaces. For a given material, the exponent α of this power law relation shows strong dependence on values of fractal dimension and RMS slope, while the coefficient β is dominated by RMS roughness. These strong correlations can be used for establishing and validating predictive models for contact stiffness on a wide range of rough surfaces.

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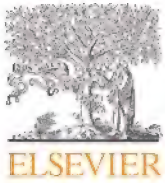
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3.2 Paper 2: Numerical modelling of contact stiffness

This second paper in this chapter presents a numerical approach for calculating the contact stiffness of fractal rough surfaces under compression. The numerical results are compared with experimental data, showing a quantitative agreement with measurements for a range of different rough surfaces. For diverse surface structures flattened by a rigid flat, a unified power-law function between the normal contact stiffness and the applied normal load can be found both experimentally and numerically. The exponent of the obtained power law relation increases with values of fractal dimension, while the magnitude decreases exponentially with the relative roughness amplitude. The numerical solutions of the proposed method are in good agreement with experimental results over a wide range of normal compression. The applicability of the presented method is further clarified in consideration of complex deformation of the compressed asperities under high stresses. Moreover, a parametric analysis is conducted to establish a correlation between contact stiffness and surface roughness descriptors. We found that the characterization method applied to determine the fractal dimension of a rough surface can affect somewhat the predicted contact mechanics. This study provides an easily implemented and computationally efficient method to connect mechanical behaviour with multi-scale surface structures, which can be utilized in design and optimization of engineering applications involving rough contacts, such as granular materials. The findings presented in Chapter 3 will be further linked with the electrical contact at rough interface presented in Chapter 4, in order to have a comprehensive understanding of the rough contact. The proposed numerical approach for rough contact along with the benchmark experiment presented in the first and second papers provides basis for modelling responses of granular materials under various stress states.

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Contact stiffness of multiscale surfaces by truncation analysis



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Nanoindentation

ABSTRACT

In this paper, we study the contact stiffness of a fractal rough surface compressed by a rigid flat plane. A numerical model based on the analysis of flat punch indentation is proposed for simulated hierarchical surfaces, which are generated using statistical and fractal descriptors collected by surface profilometry. The contact stiffness of surfaces under increasing normal load is determined on the basis of the total truncated area at varying heights. The results are compared with experimental data from nanoindentation on four types of treated rough surfaces, showing good agreement with experimental observations below a certain truncation depth. Furthermore, the limits of the model's validity are discussed by focusing on surface geometries and deformation of contacting asperities. With this proposed truncation method, we present a parametric analysis to establish a correlation between contact stiffness and surface roughness descriptors. The contact stiffness shows a unified power-law scaling with respect to the applied load over a wide range for simulated surfaces with distinct sets of roughness descriptors. The exponent of the power-law relationship is found to correlate positively to the fractal dimension while its amplitude is inversely correlated to the surface roughness amplitude. This study provides an easily implemented and computationally efficient method to connect mechanical behaviour with multi-scale surface structure, which can be utilized in design and optimization of engineering applications involving rough contacts.

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1. Introduction

The morphology of surfaces plays a determining role in interfacial phenomena including friction, adhesion, sealing, lubrication and thermal and electrical conductance [1,2]. When two solids with rough surfaces are squeezed together, the area of true contact consists of numerous contact patches of various sizes and generally comprises only a small fraction of the nominal contact area. Over the years, numerous divergent approaches have been developed to explore load dependent contact behaviour at rough surfaces, yet the scientific community remains divided regarding the reliability of their predictions [1,3–6]. The various asperity-based approaches to contact models reported in the literature can be categorized as: (1) multi-asperity contact models (i.e., statistical models), in which the heights and / or curvatures of asperities follow given statistical distributions [7–10]; and (2) surface fractality models, including multi-scale models [11–17], the boundary element method (BEM) [6] and Persson's theory [18, 19]. Further to this binary classification, the comparison between various reported models reveals discrepancies with noticeable differences between each other and with experimental results [17,19–21].

Within the above-mentioned approaches to surface structure simulation, it is also necessary to consider the contact behaviour of an individ-

ual asperity ranging from elastic, through elasto-plastic, to fully plastic deformation [8,22–29]. For purely elastic or plastic contact, the classic Hertzian model [30] and fully plastic model [31] can be applied, respectively. However, for elastoplastic regimes, individual asperity models may yield different contact responses, dominated by different deformation mechanisms [32,33]. Particularly, a self-consistent analysis was put forward by Storåkers, et al. [34] considering the general visco-elasto-plastic material indented by a spherical object. Additionally, shoulder-to-shoulder contact models for misaligned asperities were introduced to include oblique contact between pair asperities [15,35,36].

The contact behaviour of individual asperities can be combined to shed light on overall system behaviour by considering statistical and/or fractal approaches to describe surface morphologies. Pioneered by Greenwood and Williamson [7], multi-asperity contact models are based on the statistical height distribution (Gaussian or non-Gaussian), while assuming that the deformation of a given asperity is not influenced by that of neighbouring asperities. Within this framework, various implementations, considering different asperity geometries, have been developed over the past decades for the analysis of individual asperity deformation. In practice, statistical parameters for characterizing surface topography, such as variance of heights, slope, curvature, etc., have been used in this class of contact models. But these implementations assume features at a given narrow range of scales and thus depend on the

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resolution of the surface measuring apparatus and sample length [13]. However, most natural surfaces exhibit features across a wide range of length scales [37], involving diverse morphologies, which bring about complexities in the modelling of interfacial properties [3,38]. In addition, these statistical models were constructed by assuming that micro-contact forces arise principally due to deformation of asperities and are calculated considering only the base wavelength or a certain range of wavelengths, characterized by the roll-off and cut-off wavevectors of the power spectra of rough surfaces [4,12,17,19,39]. This assumption neglects the contribution of fine geometries at small length scales to the contact properties. In the fractal-theory related models, the finer surface features have been included in terms of overall contact area using an integration process [11,12,28,40].

An early appreciation for the significance of the multi-scale nature of surfaces was demonstrated by Archard [41]. In the contact theory using scale-independent parameters, Majumdar and Bhushan [42] utilized the fractal theory of Mandelbrot [43] to describe the distribution of contact areas between two rough surfaces. Yan and Komvopoulos [12] extended this theory to contact problems of three-dimensional rough surfaces, revealing the variations of the contact force and real contact area during quasi-static loading. Ciavarella, et al. [44] developed a two-dimensional fractal-based model for sinusoidal elastoplastic surfaces, and expanded the analysis to provide contact stiffness and interfacial resistance. Further relevant work was conducted by Jackson and Streater [13] using three-dimensional sinusoidal surfaces, considering the frequency spectrum of the surfaces. Pohrt and Popov [6] deduced an empirical contact stiffness model by means of the boundary element method (BEM). The validity of the method of reduction of dimensionality, by which three-dimensional contact problems are mapped onto one-dimensional elastic contacts, has been investigated for non-adhesive contact of any axisymmetric bodies [45–47]. However, the fractal dimension has been shown to change due to an applied load [38,48,49] and thus an assumption of invariant fractal dimension in fractal-theory related models may only be reasonable within a certain range of loading. Moreover, various methods [20,50,51] can be applied to quantify the fractality of a rough surface [19,20,52,53] possibly resulting in different values of the obtained fractal dimension, thus the adaptation of these methods merits further study to provide a consistent result in contact mechanics models.

The true area of contact formed between two surfaces is of prime interest and is governed by morphology, material properties, and loading conditions. The contact properties of rough surfaces subjected to an applied normal load can be, at some level, interpreted by considering the true interfacial contact area along with surface descriptors [4,46]. However, determining the real contact area between contacting bodies through experimental or numerical analyses remains challenging. Factors that influence true contact area include the asperity height, curvature, the Poisson's ratio of the material, strength and hardness and the existence of superimposed smaller asperities with similar or divergent scale-dependent properties [54]. With time, the sliding induced by expanding contacting spots [55] further affects contact morphology. Moreover, most natural surfaces exhibit features across a wide range of length scales the extent of which depends on simulation or measurement resolution [56,57].

Despite the fact that the contact area tends to present scale-dependent properties with contacting asperities in the status of incomplete contact (with non-contacting zones surrounded by the contacting ones), the truncated areas at various depths for a given resolution can be employed to represent the real contact area. This was first proposed by Abbot and Firestone [31] to describe a wear process rather than indentation or flattening. Along this line, proposed truncation models [58,59] have assumed that the contact area of an asperity pressed against a rigid flat can be approximately calculated by mathematically truncating the asperity tip. This is a reasonable assumption for small to medium interferences as within this range asperities tend to plastically deform due to the small radius of curvature. The average pressure be-

tween an asperity and a flat punch can simply be assumed to be equal to hardness, or can be related to material yield strength [7,26]. However, Jackson and Green [22] showed that this simplification results in an inverse hardening process in which the hardness actually decreases with increasing interference.

In addition to contact area the present work focuses on interfacial contact stiffness. An understanding of contact stiffness is important in contact mechanics, which plays a central role in governing the stress-dependent electrical and thermal transport between two contacting solids [6,60,61]. In the past decade, the relationship between surface structure and interfacial stiffness has been intensively studied numerically and experimentally. Numerical analyses, using methods of molecular dynamics, finite element analysis, etc., generally confirm linear proportionality between normal force and contact stiffness [39,62,63], as supported by the Greenwood–Williamson model [7] and Persson's theory [19,64]. However, other studies have reported that, for small to medium loads, the logarithm of stiffness exhibits close proportionality to the logarithm of the applied normal force [6,45,52]. In other words, the contact stiffness, k , is a power function of the normal force, F_N , as $k \propto F_N^\alpha$ (with $\alpha < 1$), which differs from the work mentioned above. Numerous experimental studies have been carried out on rough surfaces to ascertain the relationship between surface structure and interfacial behaviour under load using diverse experimental approaches and materials. Jiang, et al. [52] measured the normal contact stiffness of cast iron specimens produced using different machining methods. Wang, et al. [19] measured the contact stiffness of a rubber block squeezed against different concrete and asphalt road surfaces. Buczkowski, et al. [17] compared the normal contact stiffness determined using ultrasonic measurements with the fractal model based on Weierstrass–Mandelbrot function. Zhai, et al. [20] recently evaluated the contact stiffness at aluminium surfaces by nanoindentation tests utilizing different sized flat tips to achieve a wide range of applied stress levels. These experimental studies support the power law relationship between the contact stiffness and the applied load for certain stress ranges.

The main purpose of the paper is to propose a comprehensive contact analysis method for a three-dimensional rough surface compressed by a rigid flat. The proposed truncation method is applied to simulated fractal surface structures characterized using various statistical and fractal descriptors, to interpret the variation of contact stiffness under increasing normal loading using experimental results for reference. Iteration procedures employed in simulating fractal rough surfaces ensure a description with identical parameters which are critical in determining the normal contact stiffness. The applicability and repeatability of the presented method is discussed based on geometrical features and analyses of the studied rough surfaces. This study provides an easily-incorporated and highly-effective numerical method for predicting contact stiffness under conditions of small to medium loads. Following validation, a parametric analysis is conducted for simulated surfaces varying in surface topologies, allowing the obtained contact stiffness to be related to surface roughness descriptors. Finally, correlations between normal contact stiffness and roughness descriptors have been established and discussed with respect to fractal dimension values obtained using different methods.

2. Theoretical framework

The deformation mechanics of contacting surface asperities remains a topic of significant debate in the research community. Under most applied conditions the true contact area between rough surfaces involves only a small fraction of the nominal contact area, and for this reason the total real contact area can be considered to approximately equal to the truncation area in the present method. We consider a simple extension of the contact analysis for indentation by a flat punch [65], which reveals that there is a relation between contact stiffness, contact area, and elastic

modulus, given by

$$E_r(E_c, E_i) = \beta \frac{\sqrt{\pi}}{2} \frac{K}{\sqrt{A}}, \quad (1)$$

where K is the contact stiffness in units of N/m, A is the apparent contact area and β is a geometrical constant, taken as unity for a flat punch. E_r denotes the reduced Young's modulus including contributions from both the compressed rough surface and opposing flat surface, given by $1/E_r = (1 - \nu_c^2)/E_c + (1 - \nu_i^2)/E_i$, with the subscript i indicating the opposing surface, the subscript c indicating the properties of the compressed surface and ν being the Poisson's ratio. The contact force can be obtained through integration of the contact stiffness with respect to the indentation increment. Eq. (1) is a fundamental equation for assessing the elastic properties in indentation tests, and has been shown to be equally applicable in cases of elastic-plastic contact. If the elastic modulus is known, the relation between contact area and contact stiffness can thus be obtained [66,67].

2.1. Validation for single asperity under compression

An asperity under compression can behave elastically or plastically. For describing purely elastic contact of rough surfaces, the classic Hertzian model can be applied as suggested by previous contact models [7,26,33,42]. For a sphere under compression by a rigid plane, it is assumed that the radius of a contacting asperity, R , is very close to that of the undeformed case. In such cases, the relationship between R and the truncation microcontact radius, r' , can be obtained by $R^2 = (R - d)^2 + (r')^2$, where d is compression depth. Since R is typically orders of magnitude greater than the d , the relationship can be reduced to $(r')^2 = 2Rd$ [12,27]. In Hertzian contact, the radius of the real contact area is given by $r = \sqrt{Rd}$. Thus for a circular microcontact in the fully elastic regime the relationship between the truncation area (a') and the real elastic contact area (a_E) can be expressed as: $a' = 2a_E$.

For describing plastic deformation, Abbot and Firestone [31] developed the most widely used model for a fully plastic contact, known as the surface microgeometry model. This model assumed that the deformation of a rough surface against a rigid flat plane is equivalent to the truncation of the undeformed rough surface at its intersection with the plane. As a result, the real area of the contact can be approximated simply as the geometrical intersection of the flat with the original undeformed surface profile, and the contact pressure is the plastic flow pressure. In addition, McFarlane and Tabor [68] showed that both the normal and tangential stresses contribute to the deformation of junctions formed at the interface of contacting bodies under compression. For cases of elastic contact, the tangential stress constrains the expansion of the compressed asperity through static friction. As the contacting region between an asperity and the rigid flat is already in the plastic state of stress under purely normal loading, tangential loading, however small, may cause further yielding. Equilibrium can be maintained if the area of a contacting spot grows [68]. Therefore, for a fully plastically microcontact, the relation between the truncation area and the real plastic contact area (a_p) is: $a' \leq a_p$.

The above discussion supports that the truncation area, a' , of a single compressed asperity at any given interference lies in the range of $a_E < a' \leq a_p$, the upper and lower limits of which are determined by the contact area for fully elastic and plastic regimes, shown in Fig. 1. For a single asperity of a fractal rough surface, exhibiting a hierarchical structure, we also assume that the contact area of an individual asperity for a given surface interference will follow the relation: $a_E < a' \leq a_p$. Furthermore, the truncation method could provide contact properties between elastic and plastic behaviours. This assumption is further rationalized by comparing the dependence of contact area on contact force obtained from the concise self-consistent model for contact of inelastic materials, based on the indentation theory and von Mises isotropic flow theory [9].

For a visco-elasto-plastic contact problem, the constitutive behaviour can be simplified as $\sigma = \sigma_0 \epsilon^{m_B} \dot{\epsilon}^{n_B}$, where σ_0 is a material yield parameter and m_B and n_B are hardening and creep exponents, respectively. For a time-independent perfectly plastic material, $m_B = n_B = 0$, while in the linearly elastic case, where $m_B = 1$ and $n_B = 0$. A semi-analytical solution for the contact problem can be obtained for spherical bodies [9, 34]. In this paper, only a quasi-static situation is studied without considering creep, i.e., $n_B = 0$. Here, we consider a spherical asperity under compression by a rigid plane using the proposed truncation method and benchmark this special case to other solutions existing in the domain of contact mechanics, in Fig. 2. The maximum interference is 100 times the critical interference, $\omega_c = (\pi k H / 2 E_r)^2 R$ where $k = 0.454 + 0.41 \nu$ is the hardness coefficient of the asperity related to its Poisson's ratio, ν , and, H is the hardness of the asperity relative to its yield strength. This critical interference value marks the inception of the elastoplastic deformation [13,26,27]. The elasto-plastic solutions for materials exhibiting different hardening exponents ($m_B = 0.0, 0.10, 0.20, 0.30, 0.40, 0.50$ and 1.0) are shown in Fig. 2. The obtained numerical results from the presented truncation method (1000 truncation depth increments) lie between fully plastic and elastic regimes. Thus, to a certain extent, these results demonstrate the applicability of the truncation method for the analysis of elasto-plastic contact.

2.2. The truncation method for rough surfaces

In this work an approximate model is presented for estimating the contact stiffness of a rough surface compressed by a rigid plane with the following assumptions:

- (1) The studied fractal rough surface is flattened by the rigid plane, and contact 'islands' grow in terms of size and number through successive truncations parallel to the mean surface plane;
- (2) The truncated surface features on the rough surfaces flow plastically into the valleys of non-contacting regions;
- (3) Contacting asperities do not interact with each other;
- (4) Deformation is confined within the interface region rather than the bulk region.

In this method, a rough surface is levelled through a mountain-top removal approach in order to obtain truncation areas. For a single asperity under compression, it is a reasonable assumption to use the total truncation area, A' ($A_E < A' = \sum_{i=1}^{i=n} a'_i = A_c < A_F$, where A_c is the true contact area), at a given height to approximate the real contact area for the corresponding surface interference, ω . The contact stiffness is extracted based on the analysis of indentation tests by a flat punch [65]. Under increasing compression, smaller microcontacts expand, merging to form larger ones. Consequently, the value of contact stiffness, E_c depends primarily on the true contact area and approaches the elastic modulus of the bulk material, E . The contact stiffness (k , with the unit of N/m) can be estimated by the following expression using the true contact area (A_c), based on Eq. (1):

$$k = \beta' \frac{2}{\sqrt{\pi}} E_r' \sqrt{A_c}, \quad (2)$$

where E_r' is the (constant) reduced elastic modulus calculated for the bulk elastic properties of the tested material (Young's modulus, E and Poisson's ratio, ν) and indenter: $E_r' = [(1 - \nu^2)/E + (1 - \nu_i^2)/E_i]^{-1}$, and β' is a geometric factor of the order of unity, equalling to 1. By writing Eq. (2), we assume that the effect of surface roughness on the measured incremental stiffness can be described by considering the true contact area A_c (rather than the projected contact area A) and bulk material properties in Eq. (1). By comparing Eqs. (1) and (2), and considering that $E_i \gg E > E_c$, we obtain the following scaling relation:

$$E_c / E^* \propto \beta' \sqrt{A_c / A}. \quad (3)$$

Considering $dF = k(\omega)d\omega$, we can then obtain the loading force F through integration of contact stiffness, $k(\omega)$, during the compression

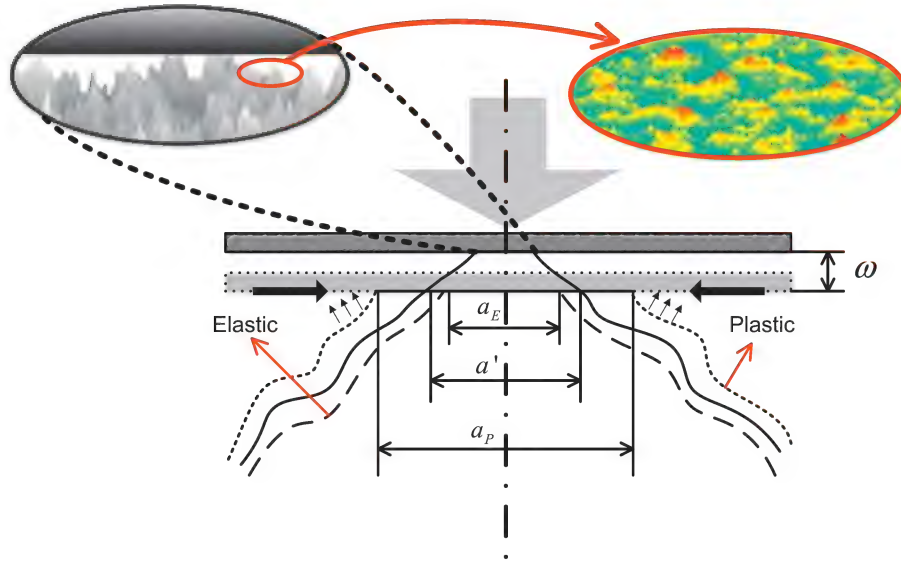


Fig. 1. Schematic of a deformed asperity under compression by a rigid plane. The areas of a_E and a_P , show the real contact areas for fully elastic and plastic regime, respectively, while a' represents the truncation area for a given surface interference of ω .

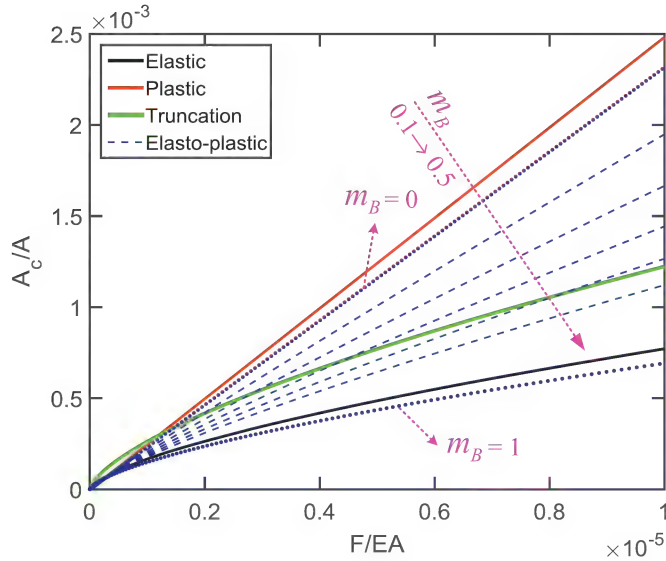


Fig. 2. Comparison of obtained contact responses of a single spherical asperity under compression of a rigid flat between the truncation model and the inelastic flattening model [34]: The normalised contact area versus normalised load is shown with A equalling πR^2 , E being the Young's modulus of the material.

with penetration increments $d\omega$. Note that we are here focusing on the hierarchical structure of the surfaces by applying normalization procedures.

3. Surface characterization and simulation

To gain insights into the validity of the presently developed truncation method for rough surfaces, we experimentally and numerically evaluated the contact stiffness for round disk samples made of aluminium alloy with surface treatments applied in order to produce distinct surface structures.

3.1. Surface fabrication and profilometry

To produce different rough surfaces, three surface treatment methods were applied: (1) polishing, (2) surface mechanical attrition treat-

ment (SMAT), and (3) abrasive blasting using various sized glass beads. More details of the surface treatment and topographical characterisation can be found in [20]. Fig. 3 shows typical scanning electron microscope (SEM) images, 3D surface topography and corresponding simulated surfaces. Over 10 digitized scans, the treated sample surfaces were characterised through descriptors of the peak-valley height R_t , root mean square roughness R_{rms} , roll-off wavelength W_r , and fractal dimension D characterized through different methods, as shown in Table 1. These roughness descriptors will be used later to generate simulated rough surfaces that have similar features for evaluating the proposed truncation method. Using the fractal dimension we are able to estimate surface structures at scales below the equipment resolution, assuming self-similar scaling in terms of geometrical features [37]. This estimation approach further improves the efficiency of the present method as utilising surface scans at sub-micron scale resolutions, such as those obtained through AFM, would be computationally intensive and would provide limited information from higher scale features.

3.2. Simulated rough surfaces

The scale-invariant parameter, i.e., fractal dimension, provides a means of describing realistic multiscale roughness. Previous studies [12,52,55] have shown that a fractal rough surface can be deterministically simulated by the Weierstrass–Mandelbrot fractal function [43], which can be written as

$$z(x, y) = W_r \left(\frac{G}{W_r} \right)^{(D_{in}-3)} \left(\frac{\ln \gamma}{M} \right)^{1/2} \sum_{m=1}^M \sum_{n=0}^{n_{max}} \left(\gamma^{(D_{in}-3)n} \right) \times \left\{ \cos \alpha_{m,n} - \cos \left[\frac{2\pi \gamma^n (x^2 + y^2)^{1/2}}{W_r} \cos \left(\tan^{-1} \left(\frac{y}{x} \right) - \frac{\pi m}{M} \right) + \alpha_{m,n} \right] \right\} \quad (4)$$

The parameter γ determines the density of frequencies used to construct the surface and, in similarity to previous work [33], is set as 1.5 based on considerations of the surface flatness and frequency distribution density. G is a height scaling parameter independent of frequency, termed topothesy [48]. The parameter W_r is the roll-off wavelength, which can be obtained from the power spectrum, determining the basic wavelength of highest scale features. In this paper, values of roll-off wavelength, W_r , of the treated surfaces are obtained by estimating the

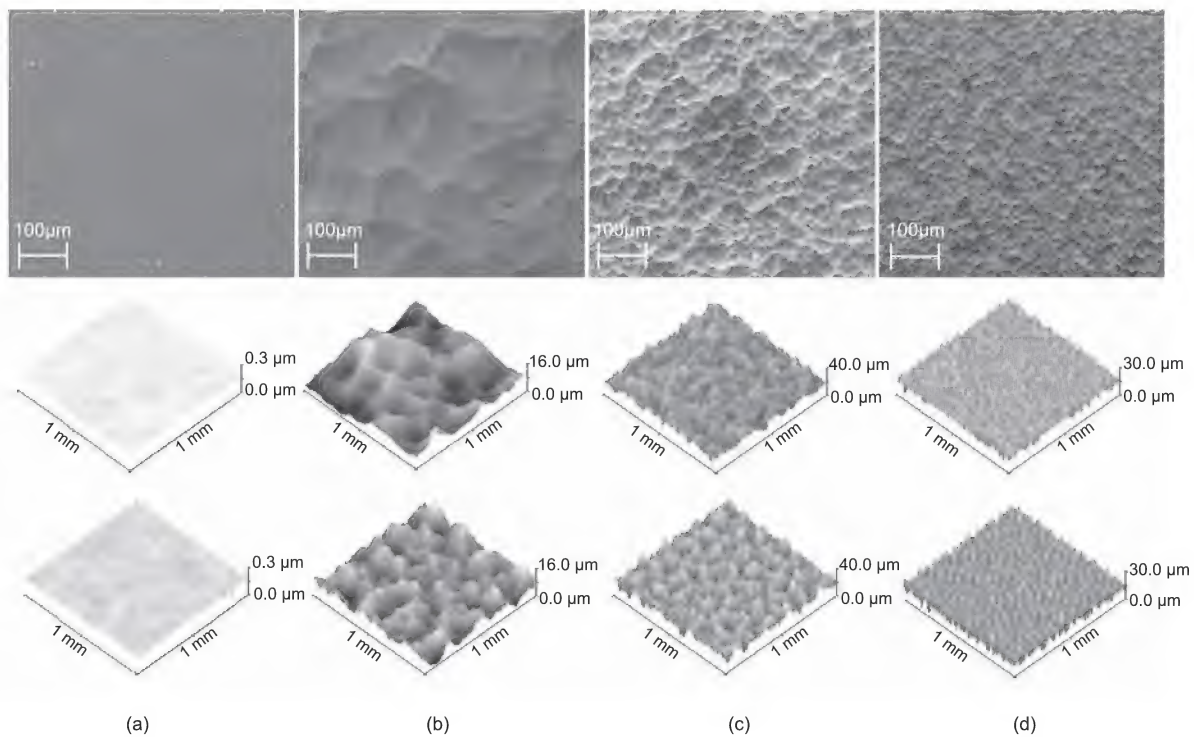


Fig. 3. SEM images, typical digitized profilometry scans (1024×1024 pixels over an area of $1 \times 1 \text{ mm}^2$) of aluminium samples subjected to different surface treatments and their corresponding simulated surfaces ($10,240 \times 10,240$ pixels) presented in row 1, 2 and 3, respectively: (a) polished; (b) SMAT with 2 mm-sized steel balls; (c) blasted with 300 μm -sized glass beads; (d) blasted with 50 μm -sized glass beads.

Table 1

Surface characterisation for digitized scans and simulated surfaces (indicated with a suffix 's' at the end of corresponding sample names).

Surface	Amplitude roughness R_t or $R_t'/\mu\text{m}$	Root mean square roughness $R_{rms}/\mu\text{m}$	Roll-off wavelength $W_r/\mu\text{m}$	Fractal dimension D_{tri}	Fractal dimension D_{box}	Fractal dimension D_{vs}	Fractal dimension D_{sp}
Polished	3.253 ± 1.939	0.057 ± 0.005	$\gg 1024$	2.093 ± 0.0618	2.024 ± 0.0410	2.103 ± 0.0628	2.317 ± 0.0788
Ps	0.263 ± 0.032	0.053 ± 0.004	$\gg 1024$	2.091 ± 0.0344	2.025 ± 0.0313	2.103 ± 0.0562	2.315 ± 0.1196
SMAT	22.184 ± 5.201	2.730 ± 0.255	~ 120	2.228 ± 0.0199	2.156 ± 0.0131	2.422 ± 0.0436	2.412 ± 0.0964
SMATs	16.210 ± 1.569	2.762 ± 0.053	120	2.225 ± 0.0232	2.161 ± 0.0297	2.417 ± 0.0487	2.421 ± 0.1222
GB300	42.376 ± 9.238	4.179 ± 0.194	~ 60	2.551 ± 0.0217	2.424 ± 0.0257	2.811 ± 0.0769	2.284 ± 0.0931
GB300s	36.865 ± 1.731	4.102 ± 0.034	60	2.555 ± 0.0425	2.417 ± 0.0316	2.813 ± 0.0521	2.282 ± 0.0876
GB50	32.239 ± 7.843	2.970 ± 0.276	~ 25	2.626 ± 0.0174	2.351 ± 0.0363	2.824 ± 0.0767	2.279 ± 0.1054
GB50s	26.239 ± 1.157	2.935 ± 0.031	25	2.625 ± 0.0328	2.351 ± 0.0395	2.825 ± 0.0876	2.284 ± 0.0933

average distances between asperities, based on surface structures measured experimentally. The frequency index n_{max} assumes finite values corresponding to the cut-off wavelength, W_c , representing the smallest distance between two adjacent pixels [12,55]. The parameter M denotes the number of superposed ridges used to construct the surface. A random number generator is used to uniformly distribute the values of random phase $\alpha_{m,n}$. Here, isotropic fractal surfaces, consisting of $10,240 \times 10,240$ pixels, with $M=10$, were generated to simulate real surfaces based on the surface descriptors obtained from digitized scans with the projective area being $1 \text{ mm} \times 1 \text{ mm}$, shown in Fig. 3.

In the fractal analysis of surface asperities, the fractal dimension and the topography as are commonly considered as two invariants. However, these two roughness parameters have been shown to vary with load [48]. In this paper, a scaling procedure based on amplitude roughness, R_t , was employed to govern the vertical properties of simulated surfaces instead of topography, G (set as 1). The values of all pixels, representing the height, were scaled according to the height-width ratio (R_t/L , where L is the side length of the surface) of the simulated surface, in order to yield the target peak-valley height. The fractal dimension of rough surfaces (for both digitized scans and simulated surfaces) was then estimated using methods of (1) triangulation [20,69], (2) box-counting

[70,71], (3) vertical sections [51], and (4) power spectrum analysis (Mitchell and Bonnell, 1990; Williams and Beebe Jr, 1993; Van Put et al., 1994; Mannelquist et al., 1998), with the obtained fractal dimension marked with D_{tri} , D_{box} , D_{vs} , and D_{sp} , respectively. The four methods employed are widely used in the calculation of fractal dimension [61,72–74], though the resulting fractal dimension may be not identical. As is shown schematically in Fig. 4, the procedure for obtaining the input fractal dimension D_{in} for simulated surfaces involves assuming an initial value of D_{in} (e.g., the value obtained from the real surface scan). A surface (1024×1024 pixels over an area of $1 \times 1 \text{ mm}^2$) is generated with inputting other required parameters, i.e., the roll-off wavelength W_r and amplitude roughness R_t . Subsequently, the calculated value of fractal dimension, D_{out} , of the generated surface is compared with the value obtained from the real surface scan and D_{in} , is adjusted accordingly. This procedure is repeated until the calculated fractal dimension, D_{out} , approaches that determined from the profilometry of real surfaces using one chosen method (e.g., error < 5%). Even though the triangulation, box-counting method, vertical sections approaches and power spectrum analysis result in different values of fractal dimension, a larger input fractal dimension consistently leads to a larger evaluated D_{out} , with all four approaches.

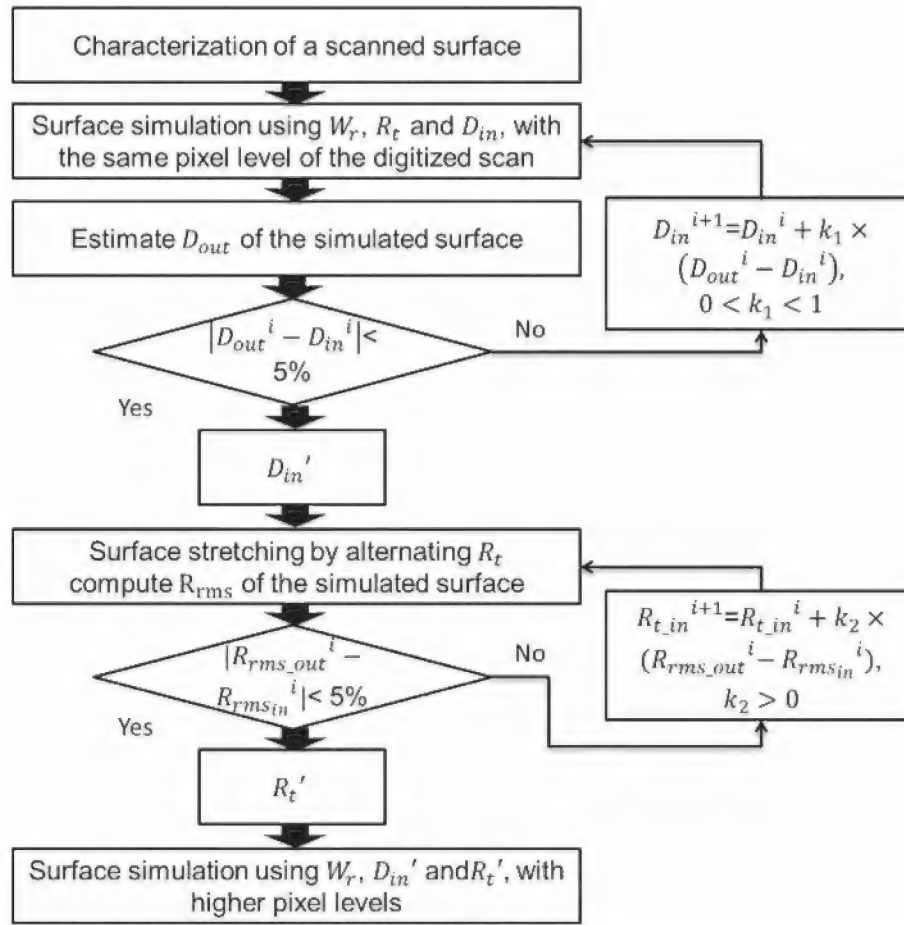


Fig. 4. Flow chart of iterative procedures for surface simulation.

As amplitude roughness R_t from digitized scans is sensitive to measurement resolution and noise, R_{rms} is employed to yield vertical features approximating real surfaces. Similar to the fractal dimension, an iteration procedure is employed to obtain a proper R_t' value with which a simulated surface can replicate the vertical geometrical features of the digitized scans. Subsequently, the obtained D_{in}' and R_t' from the iteration procedures, along with the basic wavelength, W_r , are used to generate rough surfaces at higher resolutions. The sequence of the two iteration procedures, for D_{in}' and R_t' should not be reversed, as the variation of input fractal dimension can influence the roughness amplitude, but not vice versa. The simulation of surfaces using the proposed iteration procedures are presented in Fig. 4.

The amplitude roughness, R_t , and the RMS roughness, R_{rms} , have been shown to be scale-dependent, which can be determined by the roll-off and cut-off wavelength [11]. The values of the two parameters used in the iteration procedure, shown in Fig. 4 and Table 1, are those calculated at the scanning resolution.

3.3. Influences of resolution

As the true contact area between real surfaces is difficult to definitively determine, it is important to investigate the influences of the surface resolution, as well as step sizes of truncating increments. The total contact area can be determined by summarising the contribution of contacting asperities. For a given truncation, the total truncated area, is decreasing as the resolution increases [12,43].

The contact stiffness obtained using the proposed method was studied using simulated surfaces of varied resolution. Results obtained from scans with resolution ranging from 1024×1024 to $32,760 \times 32,760$ pixels

over an area of $1 \times 1 \text{ mm}^2$ are compared in Fig. 5(a). The effects of truncation increment size are shown in Fig. 5(b). For both scenarios, the maximum truncation depths are up to half of the roughness amplitude, which is roughly the mean plane of the rough surface. The shaded error bars are obtained from ten simulations for each resolution and truncation step size. For relatively low-resolution surfaces, a clear resolution-dependence can be observed, while this trend tends to diminish as resolution increases. Similarly, simulations using 200 or more increments, up to 5000, appear to have small differences. Therefore, a resolution higher than 8192×8192 with more than 200 increment steps appears sufficient for the truncation method to provide results yielding negligible resolution sensitivity and numerical convergence.

With an increasing resolution, the contact area, and therefore, the contact stiffness approaches the asymptotic limit. This is consistent with the other fractal-based models [3,4,13,14,18], in which scale-independent contact area and contact stiffness can be achieved. Even though a converging trend has been observed based on the proposed framework, the question remains on quantitatively estimating the contact area, interfacial void, contact resistance, and any other quantity which depends on extremely fine details. The ongoing fractality shown by the hierarchical surface structures may effectively become less important for interfacial properties on a critical length scale marking the defined mechanical interaction, electron transport, heat transfer, etc. This can be generally supported by experimental research with observed values of interfacial parameters, such as contact stiffness, electrical contact resistance, and friction etc. [3,20,52,61,62,75–77]. This convergence trend will be discussed in details in our following work. In this paper, the horizontal resolution of the generated surface is set as around 100 nm, i.e., $10,240 \times 10,240$ pixels over an area of $1 \times 1 \text{ mm}^2$, and 200 incre-

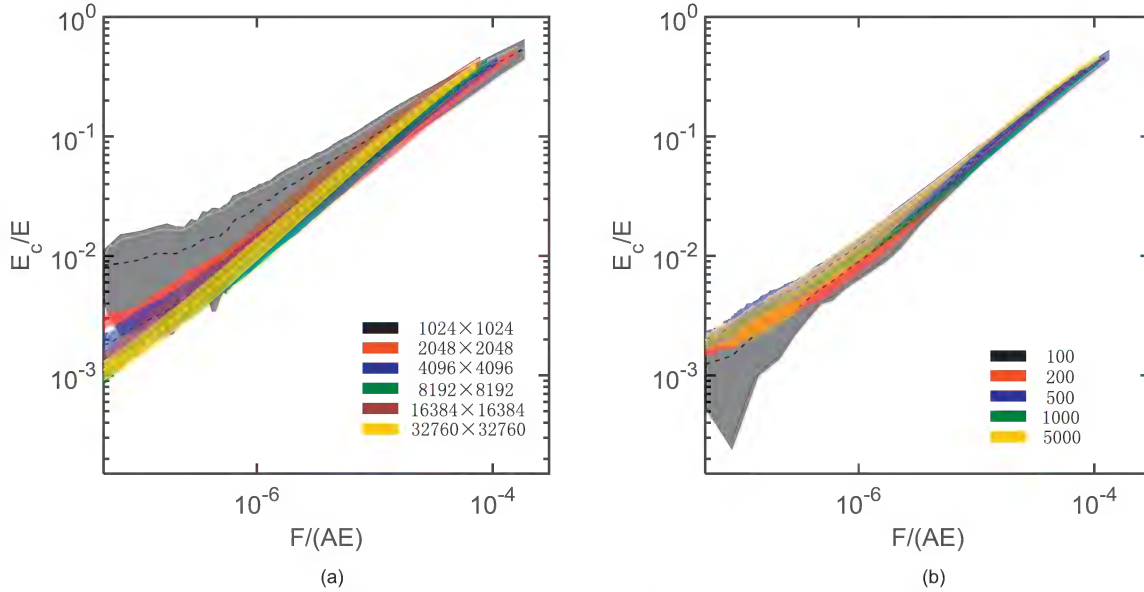


Fig. 5. Sensitivity studies on spatial resolution and increment steps on the calculated contact stiffness using the truncation method: (a) pixel level; (b) number of increment steps. The inputs of the compressed surfaces are as follows: $D_m = 2.5$, $R_t = 10,000$ nm and $W_r = 100$ μ m.

ment steps are considered in the integration process for estimating the contact force.

4. Applications and discussion

4.1. Nanoindentation on roughened surfaces

The normal contact stiffness of three types of treated surfaces was assessed using nanoindentation (Agilent G200) with three flat indenter tips of different diameters of 54.1 μ m, 108.7 μ m and 502.6 μ m (FLT-D050, FLT-D100, and FLT-D500, respectively, SYNTON-MDP). In order to evaluate only the elastic responses, partial unloading tests were successively performed at ten intervals by decreasing the applied load by 10% each time. The loading level of each successive unloading stage is twice that of the preceding one, with a maximum load of 500 mN reached during the final unloading step [20,61]. The obtained contact stiffness, in units of N/m was further transformed to surface stiffness in units of Pa through $E_c = (\sqrt{\pi}K)/(2\sqrt{A})$ where A is the projected area of the flat indentation tip. The experimental results show that the relation between the contact modulus and applied loading force can be described as $E_c/E = \beta[F/(EA)]^\alpha$. Correlations between the surface parameters and, α and β the evaluated are detailed in [20]. From the present experiments, the exponent α of the obtained power law relation shows strong dependence on surface fractality, while the roughness amplitude is found to govern the magnitude of parameter β . The maximal indentation depth reached is around 5 μ m, which is comparable with the roughness amplitude. The experimentally observed low-level compression depths suggest that the contact stiffness over a wide stress range is strongly affected by finer surface details, which are superimposed on the basic waves. These finer features may not be included in the study of contact mechanics governed by the surface structure [3–5,12]. Specifically, the considered finest feature of the roughness can be different in various engineering applications. The electrical contact resistance [61,76] and adhesion [78] have found to be sensitive to fine surface structures at smaller length scales. However, the influences of surface structures on interfacial thermal conduction can be well described using RMS surface roughness, which depends primarily on basic wavelength [79].

4.2. Comparison with experimentally measured contact stiffness

Contact analyses using the proposed truncation method are presented in Fig. 6. For SMAT and GB surfaces, the numerical results obtained from simulated surfaces are in good agreement with experimental results, particularly for low applied pressures, corresponding to small truncation depths, in the range 0 to 10.0 ~ 15.0% R_t' , 30.0 ~ 35.0% R_t' , 32.5 ~ 37.5% R_t' , and 25.0 ~ 30.0% R_t' for simulated surfaces, Ps, SMATs, GB300s, and GB50s, respectively.

It can be found in Fig. 6 that the predicted results for polished samples show the most significant divergence between numerical and experimental results in particular for interferences greater than 15% R_t' . This is likely the result of several factors, notably the misalignment of the indenter with tested surfaces and roughness features that may be present on the flat diamond tip (measured to have an RMS roughness of around 0.05 μ m) as well as adhesion, boundary effects and surface hydration, all of which yield a more pronounced effect for polished surfaces. Even though differences between the numerical analyses and experimental results can be noticed, by comparing the numerical results obtained from the mentioned models with the experimental results, we can systematically deduce the following conclusions: (1) power-law relation between the contact stiffness of a fractal rough surface and the normal load is found; (2) the roughness amplitude represented by R_t and R_{rms} plays a considerable role in determining the amplitude of the function; (3) the slope of the obtained power functions of the contact stiffness on applied normal force are in the range [1/3, 1], depending mainly on the fractal property of the surface. Comparative analyses of the experimental results with existing models can be found in [16,17,20,52,61]. It worth noting that linear relationship can be obtained within a certain range of normal compression, as shown in [17,19,64,80]. More importantly, in this paper, power-law relationships over a wide range of normal compression load have been experimentally observed and numerically demonstrated using our proposed method, covering the small-to-medium load.

4.3. Applicable truncation depth

As shown in the previous section, the proposed truncation method is capable of efficiently extracting contact stiffness on the basis of surface

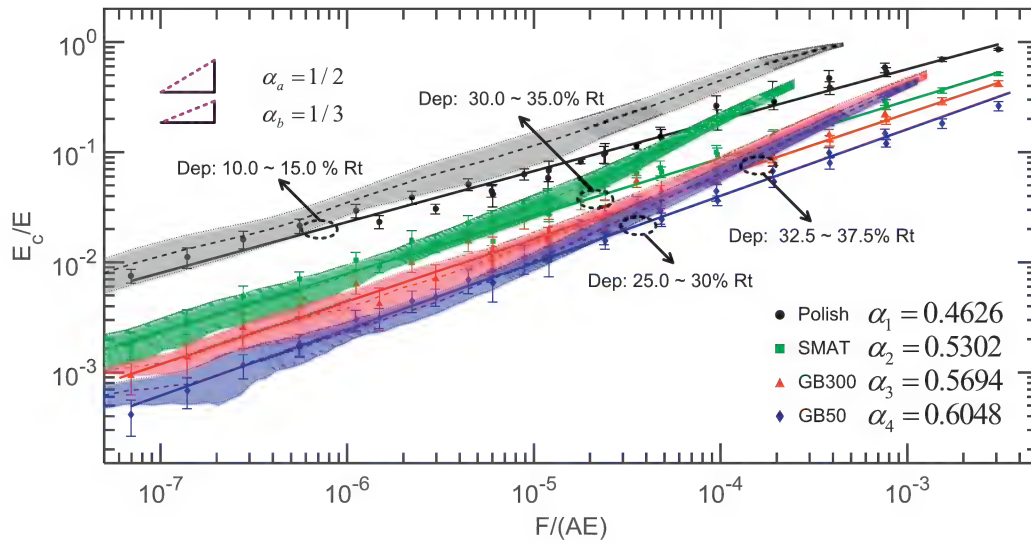


Fig. 6. Comparison of the experimental results and the contact stiffness obtained using the truncation method using D_{vs} . Normalised stiffness, E_c/E , and normalised applied load, $F/(EA)$, are used, with E being the Young's modulus of the tested material, and A the apparent contact area. The numerical results based on the present method are shown using dashed lines with shaded error bars. Solid lines (representing the fitted power functions with the exponent values provided) with error bars are for experimental results.

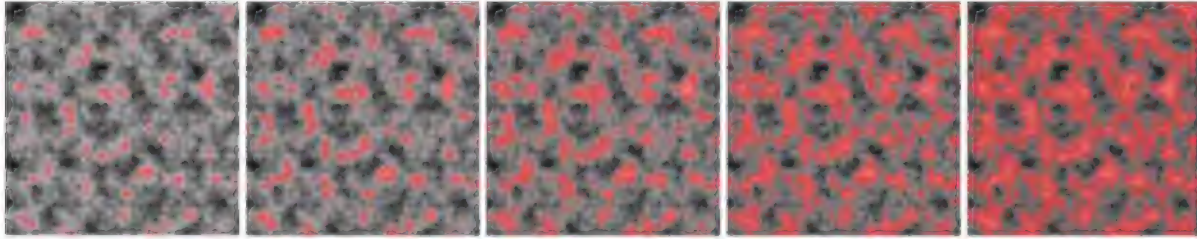


Fig. 7. Truncation sections for a typical scanned GB surface at varying heights. The truncation starts from the top of the surface with depths of 10% R_t , 20% R_t , 30% R_t , 40% R_t , and 50% R_t , and the estimated contact areas are coloured with red.

descriptors for small to medium loads. This method provides an easily implemented numerical framework to relate contact behaviour to the geometrical description of studied surfaces, with tunable resolution and calculation accuracy, meeting the requirements for various engineering applications. However, as the load and the truncation depth increase, the applicability of the presented method merits further discussion, due to the complex deformation of the compressed asperities, interaction between neighbouring asperities, adhesion, and friction. The applicable interference range is discussed below by concentrating on the surface geometries and asperity interactions.

In this model, for a given surface interference, the total contact area under compression is simply determined from numerical integration of the truncated areas of the rough surfaces, at the corresponding truncation height. As shown in Fig. 6, the proposed numerical procedure is capable of predicting the contact stiffness until the truncation depth reaches a certain value. A series of truncation sections for a typical GB300s surface are shown in Fig. 7. As the surface interference increases, small contacting spots expand, giving rise to increased interactions between asperity contact regions. A uniform rise in the height of the non-contacting regions has been experimentally observed [81], showing interactions also at asperity bases. Increasing compression can also bring about a variation in asperity shape. As the surfaces approach, contacting spots continue to grow and merge with each other, with mechanical behaviour transitioning gradually to that of a bulk material. These phenomena at large surface interferences result in limitations to the present truncation method beyond a certain level of deformation.

In order to quantitatively define the applicable range of the truncation method, we firstly consider the variation of island numbers with respect to depth, which can be employed to indicate the interaction be-

tween asperities. The numbers of contacting asperities at various truncation depths for the four types of studied surfaces over a projected area of $3 \times 3 \text{ mm}^2$ with $30,720 \times 30,720$ pixels are shown in Fig. 8(a). This size is chosen to give a meaningful statistical representation of the islands. As the interference depth increases, the number of contacting spots reaches a maximum at depths larger than those maximum truncation depths. The range of truncation depths in which the growth rate of island numbers increases monotonically coincides with the applicable range of the present method, as shown in Table 2. Furthermore, we considered the evaluation of perimeters and areas of all levelled islands at varying truncating heights using the approach proposed by Mandelbrot (1984). The correlation law between perimeter, L_T , and Area, A , for sections of truncated asperities, like islands surrounding by water, is given by:

$$L_T \propto A_T^{(D_{vs}-1)/2}. \quad (5)$$

Within a certain range, the correlation between perimeter and area follow a power law type behaviour for all the four types of studied surfaces. However, as the truncation depth further increases, the exponent experiences a downward trend as is shown in Fig. 8(b), departing from the power law behaviour shown in Eq. (5). This suggests a critical truncation depth range, in which a truncated surface can be described by an invariant fractal dimension. In this paper, at any given truncation depth, the curve fitting between L_T and A are conducted, with all obtained data for L_T and A to extract the exponent value, and the critical depth is obtained as an obvious decrease for the exponents is observed, as listed in Table 2. The exponents of the obtained power law relations are all larger than 0.5, which is the typical value of the exponent obtained in verti-

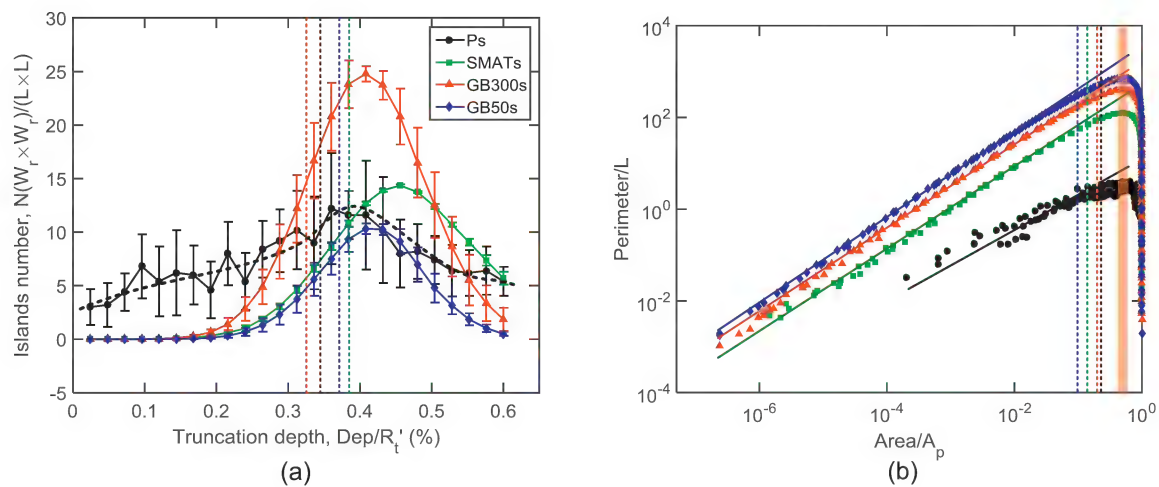


Fig. 8. Surface geometry analyses: (a) normalised numbers of contacting spots at various truncation depths for the studied surfaces, where the critical truncation depths, corresponding to maximal growth rates of island numbers, are marked with dashed lines; (b) typical log-log plots based on slit island analysis for the four types of studied surfaces (four simulations have been carried out for each type of surfaces) with the solid lines having a respective the exponent value, shown in Eq. (5).

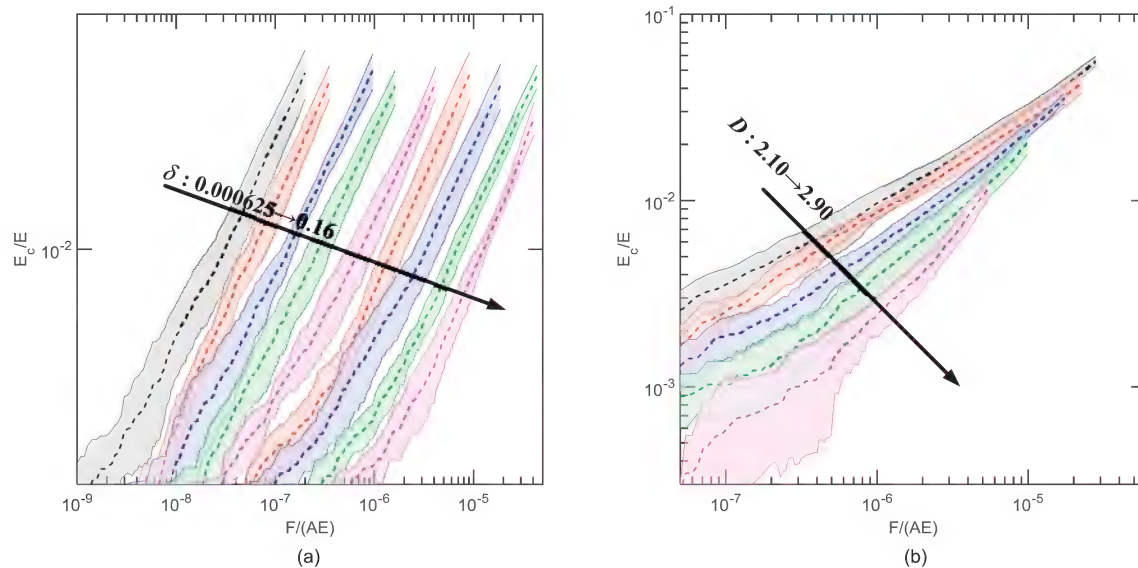


Fig. 9. Dependence of the normal contact stiffness on the normal pressure: (a) for constant fractal dimension, $D_m = 2.5$ and different relative roughness δ ; (b) constant relative roughness $\delta = 0.01$ and different fractal dimension D_m .

Table 2

Indicators for the applicable range of the proposed truncation method.

Surface	Maximum applicable depth for truncation method (% R_t)	Depth for the maximum increasing rate of island number (% R_t)	Maximum depth for surface exhibiting fractality (% R_t)
Ps	10.0–15.0	33.0–33.5	25.0–30.0
SMATs	30.0–35.0	36.0–36.5	32.5–37.5
GB300s	32.5–37.5	31.0–31.5	30.0–35.0
GB50s	25.0–30.0	34.0–34.5	27.5–32.5

cal section analysis for an asperity in regular shape, such as a sphere or cone.

Despite the fact that the two indicators we choose here are based only on surface topology, these geometrical features are closely related to the mechanical responses under normal compression. Within the range of truncation depths suggested by the two indicators, contacting asperities tend to be sufficiently isolated and thus respond to the external loading individually. Moreover, in this range, an invariant fractal dimension can be applicable to describe the multi-scale nature of the surface. At a truncation depth beyond this range, interactions between neighbouring asperities will have a significant influence on the contact, causing a

change of asperity shape, non-ignorable adhesion, sliding at interfaces etc. Even though the applicable range may also be affected by the mechanical properties of the material, including hardening, Poisson's ratio, and scale-dependent yielding strength, the purpose of this paper is to present a numerical framework to consider the structure dependent contact behaviour.

4.4. Parametric study using the truncation method

In order to relate the surface structure, represented by descriptors including fractal dimension, roughness amplitude and wavelength, to

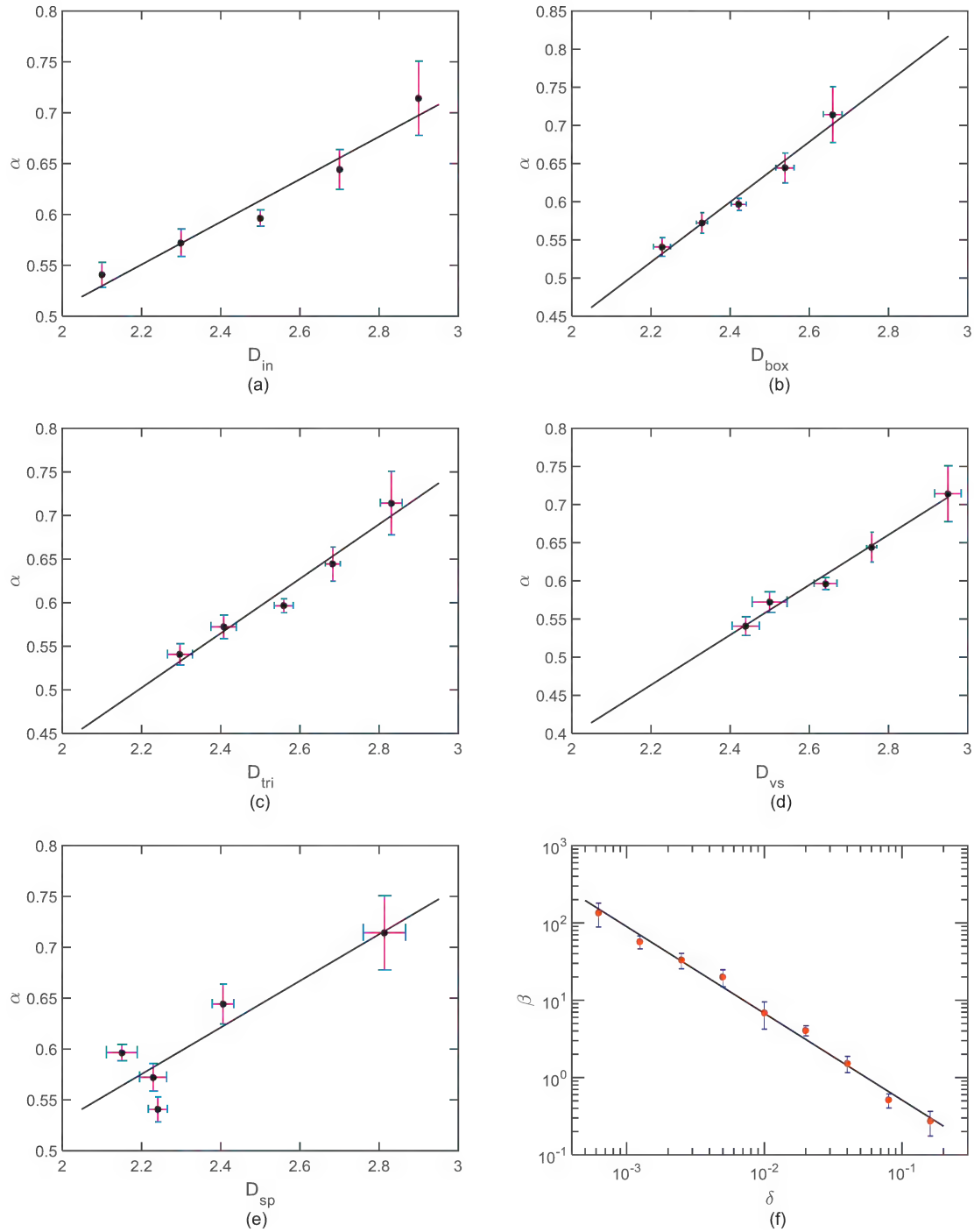


Fig. 10. (a–e) Correlation between the stiffness variation exponent α and values of fractal dimension characterized using different methods; (f) Correlation between the stiffness magnitude β and values of roughness amplitude, δ . Horizontal and vertical error bars show the corresponding standard deviations from the surface measurement and curve fitting.

contact stiffness, contact analyses are conducted for fractal rough surfaces with various surface topographies. The contact analyses were performed under conditions of small surface interferences as discussed in the previous section, in a regime where the truncation method performs well. Two groups of surfaces ($10,240 \times 10,240$ pixels over an area of $1 \times 1 \text{ mm}^2$) were generated and analysed by the present model, with the maximum surface interference being $0.1R_t$. One group of simulated surfaces was generated with a constant fractal dimension value of $D_{in} = 2.50$ and a varied roughness amplitude δ (0.000625, 0.00125, 0.0025, 0.005, 0.01, 0.02, 0.04, 0.08, and 0.16). For the second group, surfaces were set with the same relative roughness amplitude, described

as $\delta = R_t/W_r = 0.01$ and exhibited varying fractality D_{in} (2.10, 2.30, 2.50, 2.70, and 2.90). In order to further test the repeatability of the present model, we analysed five simulated surfaces for each individual set of input roughness parameters. Fig. 9 shows that the predicted contact stiffness exhibits a power-law relation with the applied normal load, which can be described as $E_c/E = \beta[F/(EA)]^\alpha$. The parameters of exponent α and magnitude β are related only to surface structure. The exponent α increases with values of fractal dimension, D_{in} , while the relative roughness amplitude, δ , indicating primarily the vertical scale of the rough surface, dominates β . The correlations for the stiffness parameters, i.e.,

Table 3
Fitting functions for the stiffness parameters, α and β , using D_{in} and δ .

Fractal dimension	α	Coefficient of correlation (for α)	β	Coefficient of correlation (for β)
D_{in}	$\alpha = 0.209D_{in} + 0.090$	0.940	\	
D_{tri}	$\alpha = 0.313D_{tri} - 0.186$	0.948	\	
D_{box}	$\alpha = 0.394D_{box} - 0.347$	0.966	\	
D_{vs}	$\alpha = 0.328D_{vs} - 0.258$	0.980	\	
D_{sp}	$\alpha = 0.229D_{sp} + 0.071$	0.741	\	
δ	\		$\beta = 0.0386 \delta^{-1.122}$	0.992

α and β , using δ and D were obtained and shown in Fig. 10. The corresponding fitted functions are presented in Table 3.

The plots shown in Fig. 10 examine the effects of using different methods for the evaluation of the fractal dimension of rough surfaces in contact [3,6,11–13,20,61], namely the triangulation method (D_{tri}), box-counting method (D_{box}), vertical sections method (D_{vs}), and power spectrum analysis (D_{sp}). The obtained fractal dimensions are expected to differ somewhat due to the use of different calculation methods. As shown in Fig. 10 and Table 3, using fractal dimension obtained from different methods will produce different fitting functions in terms of $\alpha(D)$. Thus it is necessary to clarify how the fractal dimension is extracted and the applicable interference and/or load range of the chosen approach of contact mechanics. However, the regression factors for fitting functions, $\alpha(D)$, are greater than 0.94 in most cases, except the one using the power spectrum analysis, demonstrating a strong correlation with fractality.

5. Conclusion

In this paper, we propose an efficient numerical approach for calculating the contact stiffness of fractal rough surfaces under compression by considering a series of truncation sections and discuss the applicable range of this method. Numerical predictions of contact stiffness are benchmarked with experimental data over a wide range of applied normal loads, showing a quantitative agreement with measurements for a range of different rough surfaces. Parametric analyses were carried out for simulated surfaces with varying surface topologies, to establish correlations between the obtained contact stiffness and surface roughness descriptors, including the fractal dimension and roughness amplitude. Based on the results and discussion presented, the following conclusions can be drawn:

- (1) Fractal rough surfaces can be well described using three key parameters including roll-off wavelength, amplitude roughness and fractal dimension. For diverse surface structures flattened by a rigid flat, a unified power law function between the contact stiffness and the contact load can be found both experimentally and numerically. The exponent of the obtained power law relation increases with values of fractal dimension, while the magnitude decreasing exponentially with the relative roughness amplitude.
- (2) The numerical solutions of the proposed method for contact stiffness versus contact load are in good agreement with experimental results over a wide range of normal compression. The surface interference range over which the proposed truncation method is applicable, can be determined by monitoring the variation of the number of contact islands in truncated sections, and the evolution of the total perimeter and area of these islands, as truncation deepens. These geometrical features are closely related to the mechanical responses under normal compression indicating the interactions between asperities. Within the applicable range, an invariant fractal dimension can be employed to present the hierarchical properties of the surface structure.
- (3) The parametric analyses presented in this study suggest that different methods for characterizing the fractal dimension of a rough surface may lead to different predicted contact stiffness within the applicable interference for approaches based on fractal theory. This implies that the characterization method applied to de-

termine this surface descriptor can affect somewhat the predicted contact mechanics.

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4 Electrical-mechanical Behaviour at Rough Interfaces

Electrical contact is a complicated phenomenon due to the multi-scale surface structures and many involved mechanisms of resistance modification, including dielectric breakdown, localized current-induced welding, percolation collective process, and arising chemical reactions. The understanding the involved multi-physics coupling between electrical, thermal and mechanical fields is of considerable importance in quantifying the single contact resistance at a rough interface and the overall contact macroscopic resistance throughout the granular media.

4.1 Paper 3: Electrical contact resistance at rough interfaces

This paper presents an experimental investigation on the electrical contact resistance between rough surfaces in close contact under various compressive stresses. We considered isotropically roughened aluminium disks with upper and lower surfaces modified through polishing and sand blasting using different sized glass beads. Fractal geometry and roughness descriptors, including root mean square values of roughness and slope, were used to describe the topography of sample surfaces, based on the digitized profiles obtained from interferometry-based profilometry. The electrical contact resistances at the interfaces were measured through various scenarios realised by controlling experimental parameters including testing time, testing current and mechanics loading. The experimental results show that the measured resistance depends closely on the measurement time, testing current, surface topology, and mechanical loading. For properly chosen measurement time and testing current, we demonstrated that a power-law relation exists between the measured electrical resistance and normal stress across a certain range, with the absolute values of exponents rising with the fractal dimension of the surfaces. The work will be of interest to a general community in contact mechanics, as well as in material engineering. The observed evolution of electrical contact resistance at various loading conditions helps to understand the electrical transport in granular materials.

This paper has been published in *ASCE Journal of Engineering Mechanics* in 2015. I was the primary researcher and author of this paper, being supervised by Dr. Yixiang Gan and Dr. Dorian Hanaor.

Stress-Dependent Electrical Contact Resistance at Fractal Rough Surfaces

Chongpu Zhai¹; Dorian Hanaor²; Gwénaëlle Proust³; and Yixiang Gan⁴

Abstract: The electrical contact resistance between contacting rough surfaces was studied under various compressive stresses. The samples considered here were isotropically roughened aluminium disks with upper and lower surfaces modified through polishing and sand blasting using different sized glass beads. Fractal geometry and roughness descriptors, including root mean square values of roughness and slope, were used to describe the topography of sample surfaces, based on the digitized profiles obtained from interferometry-based profilometry. The electrical contact resistances at the interfaces were obtained by applying a controlled current and measuring the resulting voltage, through the following scenarios: (1) over time for various applied testing currents, the resistance relaxation curves were measured at constant loads; (2) through voltage-current characteristics by means of a logarithmic sweeping current, the influence of the testing current on the electrical response of contacting rough surfaces was evaluated; and (3) for a given testing current, the electrical resistance through interfaces of different surface structures was measured under increasing compressive stresses. The experimental results show that the measured resistance depends closely on the measurement time, testing current, surface topology, and mechanical loading. At stresses from 0.03 to 1.18 MPa, the electrical resistance as a function of applied normal stress is found to follow a power law relation, the exponent of which is closely linked to the surface topology. DOI: [10.1061/\(ASCE\)JEM.1943-7889.0000967](https://doi.org/10.1061/(ASCE)JEM.1943-7889.0000967). © 2015 American Society of Civil Engineers.

Author keywords: Rough surfaces; Electrical contact resistance; Branly effect; Fractal dimension.

Introduction

The properties of the electrical contact resistance (ECR) at electrical connections are of tremendous importance in many engineering applications, including resistance spot welding, diagnostic tribology, signal, and current transmission (Crinon and Evans 1998; Kogut and Komvopoulos 2003; Kogut and Komvopoulos 2005; Slade 2013). For most applications, e.g., electronic connectors (Bryant and Jin 1991), switches, and electrode structures of batteries (Meulenberg et al. 2003), a low and stable contact resistance is sought after. In these situations, numerous factors can affect the current flow through contacting surfaces, e.g., surface topography, environmental conditions, mechanical loads, the presence of a coating layer, and applied voltage. Vibrations, fretting, thermal shock, and chemical contamination are principal failure mechanisms at electrical contacts often resulting in the malfunction of electric circuits. Effective electrical contacts require reliable mechanical contact in order to avoid failure. Surface characteristics, such as roughness, curvature, and fractal dimension, play an important role in facilitating the closing and opening of electric circuits created by the close contact between surfaces (Bryant and Jin 1991; Oh et al. 1999; Kogut and Etsion 2000; Falcon et al. 2004; Kogut 2005).

The true area of contact at an interface is considerably smaller than the apparent or nominal contact area because of the existence of surface roughness (Archard 1957; Bowden and Williamson 1958; Greenwood and Williamson 1958). When two rough surfaces are squeezed together contact is made through individual asperities with contact patches that can extend in size down to the nanoscale. These contact junctions exhibit electrical and mechanical properties that may diverge from bulk properties (Jackson et al. 2012). Current flowing through rough interfaces is scattered across many contacting asperities with electronic transport involving multiple mechanisms, including quantum tunneling (Yanson et al. 1998; Foley et al. 1999; Agrat et al. 2003), Sharvin contact (Sharvin 1965), and Holm contact (Holm and Holm 1967), depending on the size of contacting junctions and the mean free path of electrons. Early work by Holm and Holm (1967) concluded that the ECR is affected by both the constriction resistance resulting from the limited areas of true contact at an interface and interfacial resistance due to the inevitable presence of resistive surface films, such as oxide layers. When two metal surfaces are compressed together with sufficient pressure, surface asperities can penetrate the oxide layer thus forming metal-to-metal contact patches. When the size of contacting asperities becomes larger than the mean free path of electrons, Holm contact will be the dominant transport mechanism, resulting in a relative low resistance. This concept has been further developed by Chang et al. (1987), Ciavarella et al. (2004, 2008), and Jackson et al. (2009). These theoretical and computational studies assumed that individual microcontacts formed by asperities, where the circuit continuity was established, governed specific electron transport regions, limited by the measuring resolution.

The asperities of contacting surfaces tend to exhibit complex geometries and structures at a wide range of length scales, governing physical properties and interfacial phenomena and giving rise to constriction resistance. The fractal topography based on scale-invariant parameters provides an effective means for modeling engineering surfaces with random self-affine multiscale

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properties (Yan and Komvopoulos 1998; Kogut and Komvopoulos 2004). A general ECR theory based on fractal geometry was proposed in order to describe the effects of contact loads, elastic-plastic deformation of the contacting asperities, surface topography, and material properties on size-dependent electrical resistance of the microcontacts comprising the true contact area (Mikrajuddin et al. 1999; Kogut and Komvopoulos 2003). Kogut et al. (2005) further investigated conductive rough surfaces separated by a thin insulating film. Semiempirical power-law type correlations between the contact resistance and the normal pressure have also been proposed. For contacting surfaces separated by superficial oxide or impurity layers, the power law is found, with the exponent value possibly greater than 1 (Milanez et al. 2003; Falcon and Castaing 2005; Paggi and Barber 2011). The power-law relation between the applied normal load and the contact conductance at rough surfaces has been reported on the basis of the incremental stiffness, which was found to be linearly proportional to the contact conductance (Barber 2003; Pohrt and Popov 2012; Pohrt and Popov 2013; Hanoar et al. 2015).

Furthermore, the influencing factors of ECR include the physical and chemical origins and details of the oxidation processes in corrosive environment (Sun et al. 1999; Sun 2001), surface diffusion (Crinon and Evans 1998; Ogumi and Inaba 1998) and fretting corrosion (Bryant 1994). Thin oxide or hydroxide layers, acting as resistive films with typical thickness ranging from 1 to 10 nm, tend to form at metallic surfaces, covering the contacting surface, and increasing the contact resistance (Bryant and Jin 1991). In conduction through interfaces, insulating layers can bring about high electrical resistance under conditions of low current. With an increasing current this high initial resistance decreases by several orders of magnitude showing nonlinear conductivity phenomena, which is called the Coherer effect or Branly effect. The process is featured by voltage creep, hysteresis loops, and voltage saturation effects (Castaing and Laroche 2004; Falcon and Castaing 2005; Bourbatache et al. 2012; Tekaya et al. 2012). Many mechanisms of resistance modification have been reported in conduction phenomena through rough interfaces, including electron tunneling through oxide layers and voids (Creysseles et al. 2007), dielectric breakdown of oxide layers (Dorbolo et al. 2002), localized current-induced welding (Falcon and Castaing 1993), percolation collective process (Falcon and Castaing 2005), and chemical disorder arising with random composition (Creysseles et al. 2009).

Despite recent progress of nanoscale testing technology and surface morphology characterization, a stress-dependence of electrical conduction behavior through rough interfaces with random multiscale texture remains largely unknown. Particularly, existing

experimental results are limited. In this paper, the measured ECR of aluminium disk stacks loaded in compression is presented. The surfaces of the disks were modified by polishing and sand blasting in order to obtain a range of rough surfaces. Compressive loads, testing current, and time were varied to study their influence on the ECR.

Sample Preparation and Surface Characterization

In this paper, round disks, with a diameter of 25 mm, made of aluminium alloy were used as specimens. Surfaces were polished followed by sand abrasive blasting processes using different sized particles to modify the surface details and structures at various scales (Hanoar et al. 2013). For each individual sample, both top and bottom were equally treated through standard polishing and sand blasting procedures. The average diameters of the two selected groups of glass beads used in blasting treatments were 50 and 300 μm . Fig. 1 shows scanning electron microscope (SEM) images of the different surface types used in this work. Samples that have been blasted using glass beads of 50 μm are found to exhibit the most complex and roughest surface texture. The treated aluminium surfaces were scanned using an optical surface profilometer (NanoMap 1000WLI, AEP Technology, California) to obtain three-dimensional digitized topographies. Subsequently, RMS roughness, RMS slopes, and fractal dimension were utilized to characterize and compare the surface geometries.

As shown in Table 1, three types of samples with distinct surface details were prepared and characterized prior to electrical testing, with S1, S2, and S3 representing the polished samples, samples blasted using 300 μm diameter glass beads, and samples blasted using 50 μm diameter glass beads, respectively. Before the standard sand blasting treatment, all the sample surfaces were polished and prepared using several polishing steps with the final step using 1 μm diamond suspension. Then, the processed samples were properly cleaned by water and compressed air to remove any embedded glass beads. Cleaning with ethanol and heat treatment (from 110 to 120°C) were also applied to remove surface contamination and moisture. For each surface, the mean values of roughness descriptors with standard deviations over ten 1×1 mm scans from different samples are shown in Table 1. The scaled triangulation method (Dubuc et al. 1989; De Santis et al. 1997; Zahn and Zösch 1999) was used for the calculation of fractal dimension values. A power law relationship is found between the calculated surface area and the length resolution of digitized scan of all the surfaces, exhibiting self-affinity over a range of length scales (from 1 to 100 μm).

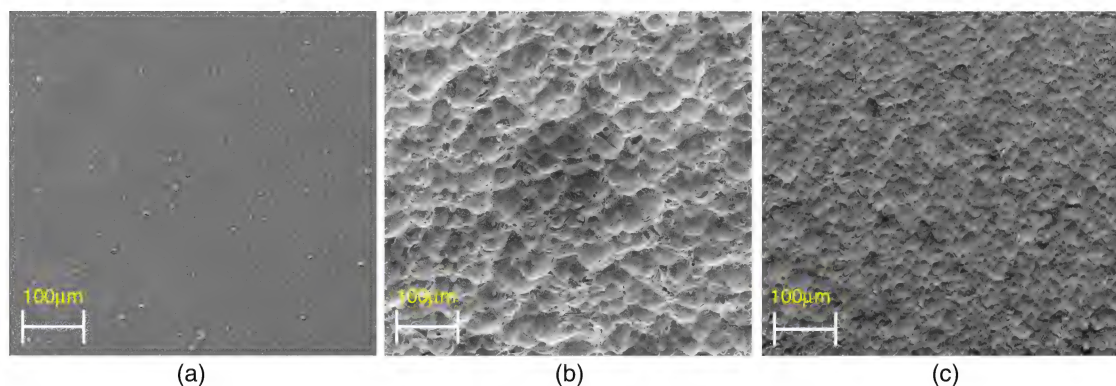


Fig. 1. SEM images of aluminium samples with different surface treatments: (a) polishing treatment; (b) blasted with 300 μm diameter glass beads; (c) blasted with 50 μm diameter glass beads

Table 1. Sample Surface Characterization with Different Treatments

Sample type	Surface treatment	RMS roughness $R_{RMS}/\mu\text{m}$	RMS slope R_s	Fractal dimension D_f /triangulation
S1	Polished	0.057 ± 0.005	0.009 ± 0.001	2.093 ± 0.0620
S2	Blasted by glass beads of $300\ \mu\text{m}$ diameter	4.179 ± 0.194	0.224 ± 0.015	2.551 ± 0.0217
S3	Blasted by glass beads of $50\ \mu\text{m}$ diameter	2.970 ± 0.276	0.202 ± 0.010	2.626 ± 0.0174

Surfaces blasted with glass beads of $300\ \mu\text{m}$ diameter reveal the highest RMS slope and RMS roughness. The lowest values of RMS roughness, RMS slope and fractal dimension are found for the polished surfaces. The values of lower-roll-off wavelength in the power spectrum of all of the surfaces used were found to be $\sim 100\ \mu\text{m}$, which is remarkably smaller than the disk diameter (25 mm).

Results and Discussion

Experimental Setup

For each type of sample with similar surface characteristics, electrical resistances were measured for stacks of 11 samples, giving 10 rough to rough interfaces, by means of a source/measurement unit (Agilent B2902A, Keysight Technologies), under various compressive loading forces, as shown in Fig. 2. In this experimental setting, the resistance formed by 10 specimen interfaces placed between two polished plates of the same material instead of one single interface is measured, aiming to achieve a higher precision, larger linear range, and better robustness against measurement noises from the connecting wires, loading device, and measurement unit. A stack of samples can also diminish effectively the experimental errors from the variation of samples with the same surface treatment. Before the experiment, the sample stack was aligned by rotating the top pressure head of the loading machine and using a piece of rubber placed between the pressure head and the top polished plate.

Prior to the measurement of stress-dependent resistance, the following tests were performed: (1) resistance creep tests and (2) sweeping current tests, to exclude the influences from the applied current and measurement time. In the following sections, the results of these tests will be presented and discussed. Finally,

the stress-dependent ECR was measured using the applied current of 10 mA and measuring time of 0.01 s at each individual stress level.

Resistance Creep Tests

The relaxation dynamics of resistance for a given constant current were first investigated to find out the effect of current on the measured resistance over time. The resistance degradation curves at various applied testing currents of 10, 20, 80, 320, and 1,280 mA were recorded, under an applied pressure of 0.061 MPa, as shown in Fig. 3(a). Then, a constant current (320 mA) was applied to study the resistance relaxation under various applied compressive stresses, ranging from 0.031 to 0.490 MPa, as shown in Fig. 3(b). Multiple tests were performed for each individual loading condition, but for clarity here not all the experimental data collected were shown and the trends shown in Fig. 3 are true for all the data collected. The measured resistance under various loads and current conditions gradually decreases with respect to time. For tests carried out under a constant normal load, shown in Fig. 3(a), a higher current brings about a more significant decrease in measured resistance. More specifically, the highest current of 1,280 mA over 200 s was found to cause a decrease of around 15% with reference to the initial measured resistance, whereas a variation of less than 1% was achieved at the lowest current (10 mA). For the tests with the same testing current (320 mA), a higher pressure reduces the trend, which is presented in Fig. 3(b). The measured resistance of the samples under an applied pressure of 0.490 MPa exhibited a slight decrease of less than 2% with reference to the initial resistance, whereas a drop of approximately 15% was found under an applied pressure of 0.031 MPa. For all the tests when the conduction time is less than 1 s, the decline of the measured resistance due to the current is less than 0.1%.

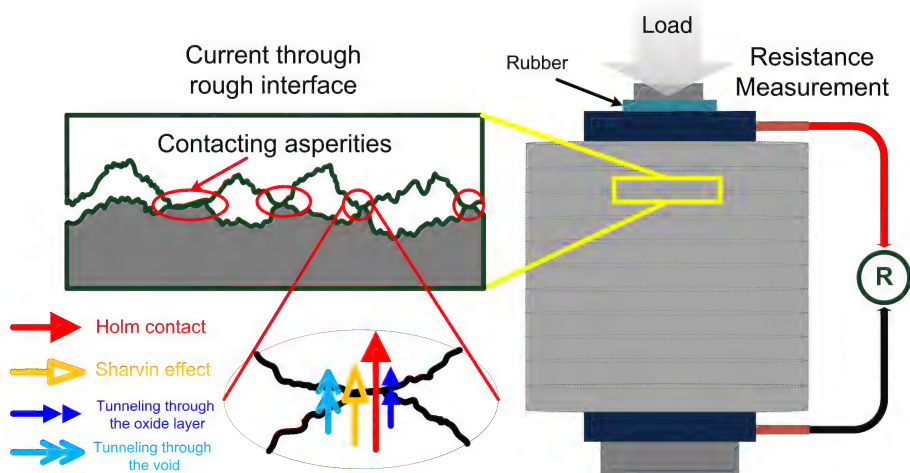


Fig. 2. Experimental setup for the resistance measurement of a stack of rough surfaces and schematic of the current flow through rough interfaces by means of Holm contact, Sharvin contact and quantum tunneling

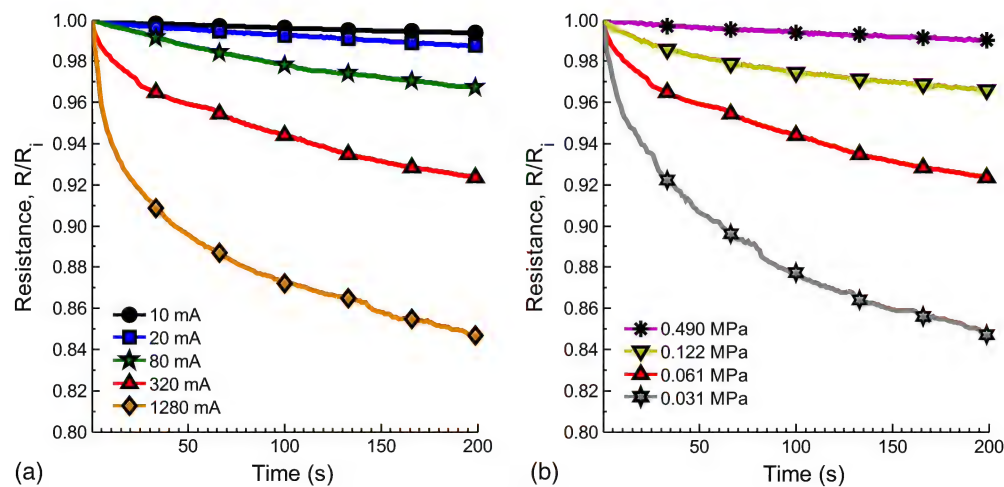


Fig. 3. Typical resistance relaxation curves over 200 s, with R_i being the initial measured resistance: (a) with constant applied stress being 0.061 MPa under various testing current (10, 20, 80, 320, and 1,280 mA with the initial resistance being 1.57, 1.46, 0.93, 0.76, and 0.23 Ω , respectively); (b) with the constant testing current being 320 mA under various compressions (0.031, 0.061, 0.122, and 0.490 MPa with the initial resistance of 2.01, 0.76, 0.59, and 0.18 Ω , respectively)

Sweeping Current Tests

The influence of the testing current on measured ECR was assessed by sweeping current tests. Here the applied sweep current test consisted of two phases with the *loading* phase (P1) having a logarithmically increasing current from 0.0001 to 1.5 A and the *unloading* phase (P2) having a logarithmically decreasing current from 1.5 A back to 0.0001 A. During the sweep procedure, the voltage was recorded at a data acquisition frequency of 2 kHz corresponding to the electrical current imposed. The normal pressure applied on the measured samples was 30 N, corresponding to a stress level of 0.061 MPa. The sweep process, including both phases, was accomplished within 0.2 s in order to avoid the occurrence of electrical degradation over time. As discussed in the previous section, when the conduction time is shorter than 1 s, the time-dependant reduction in resistance is negligible. Figs. 4(a and b) show the measured voltage and contact resistance with respect to testing current for the three types of samples. The shape of voltage hysteresis loops

is seen to be consistent for the three surface types while presenting different voltage levels. In Fig. 4(b), the measured results of all three surfaces demonstrate similar trends known as the Brany effect (Castaing and Laroche 2004; Falcon and Castaing 2005), i.e., the measured resistance begins to drop after the testing current reaches a certain value, after which the resistance appear to be irreversible as the current sweeps back. The corresponding threshold current values for S1, S2, and S3 are around 150, 80, and 60 mA, respectively, and the values seem to inversely correlate with the fractal dimension of the surface. However, the definite correlation between the RMS roughness and the threshold current can only be concluded by extending the types of surfaces. The use of RMS roughness is limited in predicting interface behaviors, whereas the fractal dimension, a cross-scale descriptor, is extensively used to interpret surface phenomena, such as conduction properties at rough interfaces (Kogut and Komvopoulos 2005; Persson 2006). The transition at the threshold current may come from the electrothermal coupling of the microcontacts at rough interfaces.

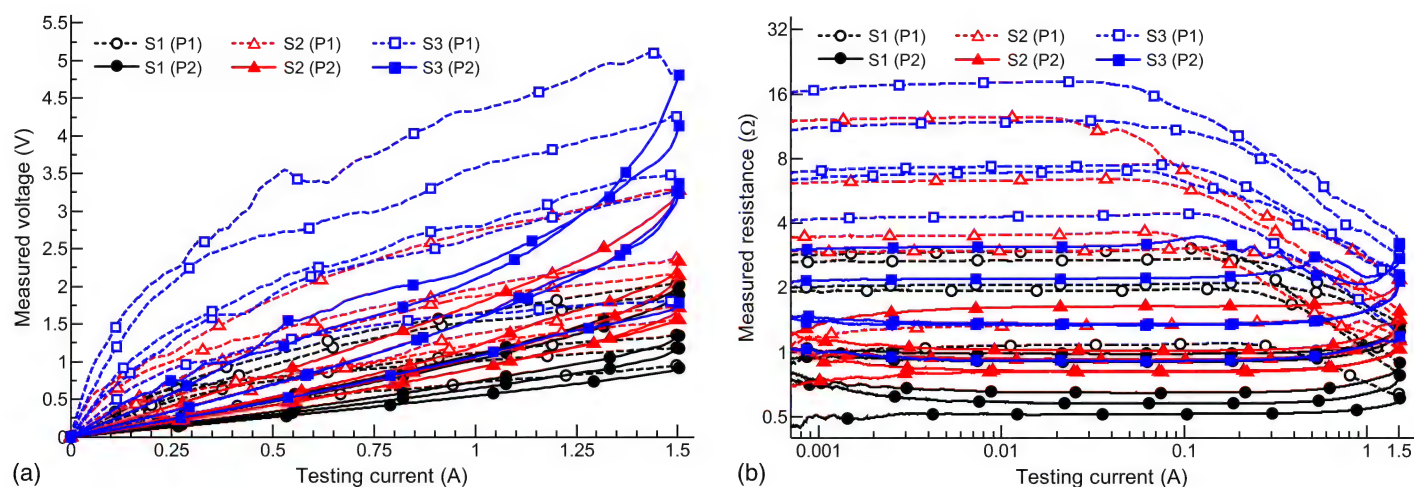


Fig. 4. Typical measured results for three types of samples using sweeping currents under a constant stress of 0.061 MPa; the data presented by the solid lines describes the first phases with increasing testing currents and the dashed lines show the second phases with decreasing currents: (a) variation of the measured voltage; (b) resistance-current characteristics

For testing currents higher than approximately 5 mA and lower than the threshold current values, the measured resistances remain stable on two plateaus in both P1 and P2, with a larger value obtained for P1, shown in Fig. 4(b). In other words, ohmic behavior at low levels of electrical current was found for all three types of surfaces. As shown in Fig. 4(b), at low testing currents (lower than 1 mA) in either P1 or P2, the measured resistances exhibit instability, especially for polished samples. Measurement noise for all three surfaces is observed at similar levels, which are amplified in logarithmic scale, being prominent when the measured resistances are small.

For each type of surfaces, the range of measured voltage and resistance was found to vary in five different tests under the same experimental conditions, e.g., the measured resistance of polished samples spreads from 0.5 to 3 Ω . From the experimental results, the surfaces described by higher fractal dimension demonstrate larger values of measured resistances. Specifically, the smallest resistances are observed from polished samples and the largest resistances are measured from the samples blasted using glass beads of 50 μm diameter. The polished samples present the smoothest and the most stable trends among the three types of surfaces. High level noises are found for surfaces after sand blasting procedures showing more complex topographies, which may cause the sensitivity to the microvibration from the loading device and the electrical noise from the measurement unit.

By rearranging the sample stacking order or by rotating the samples to achieve different contact configurations one can retrieve the voltage hysteresis loops and repeat the processes depicted in Fig. 4. Despite this, all the samples used were repolished and sand blasted again between successive tests to avoid the accumulation of surface modification by current flow as the sweep procedure may bring about some localized modification of the surface characteristics or possible changes of surface chemical composition.

A further test was done by driving successive back-and-forth scanning current cycles with increasing current range. A typical obtained result for polished samples is shown in Fig. 5. Here each successive current cycle was of 0.2 s duration and compressive pressure was maintained at 0.02 MPa during all nine sweep cycles. The sweeping current range was increased by a factor of $\sqrt{2}$ at each consecutive cycle until the current sweep reached 1.5 A. There exists a pause of around 10 s for data exporting and setting of

the source/measurement unit. Additionally, tests with a longer pause up to several minutes were conducted and presented the same trends. For all nine cycles, the electrical current flows at different levels can modify the ECR of interfaces at different degrees. At each individual cycle, the measured voltages and resistances tend to be reversible when the testing current is lower than the maximum current of the previous cycle, i.e., the measured voltages and resistances in P1 branch are inversely along the track of P2 of the previous cycle, exhibiting ohmic conduction properties. The envelope line constituted by P1 of the first cycle, P2 of the last cycle, and the resistance declining segments in P1 of all nine cycles, exhibits a similar curve shapes as that of each individual cycle. The threshold current values at the turning point for measured resistance in all cycles grow from around 100 mA to 1 A, along with the increasing range of the imposed sweeping current.

Even though all the tests were performed over a short duration, i.e., 0.2 s, the Joule heating generated from the testing current may still have an effect on the measured resistance. In Fig. 5(b), evident upward and downward trends are observed at the end of P1 and at the beginning of P2, respectively. The aluminium has a lower heat capacity than the oxide. The Joule heating is likely to contribute to the trends to a certain extent. The irreversible Joule heating from the injected current, which accumulates with respect to time, cannot fully explain the reversible upward and downward trends at the end of P1 and the beginning of P2. Another possible reason can be the charging and discharging processes occurring during the tests. The contact of rough interfaces can be regarded as a complex network of resistors and capacitors that vary with the applied pressure and injected current. Moreover, this process is very similar to the conduction behaviors in semiconducting devices showing nonlinear and reversible properties.

A further experimental study was conducted in order to shed light on relations between the applied load and ECR. For polished surfaces, five typical voltage-current loops and resistance-current characteristics under various loads ranging from 0.031 to 0.490 MPa were obtained and are shown in Fig. 6. Under a high compressive pressure (more than 0.490 MPa), the loops become flat, i.e., P2 follows the same path as P1. The Branny effect tends to be harder to capture when the pressed surfaces were in contact at sufficiently high stress values. A high level of applied stress leads to better stability and repeatability of ECR measurements.

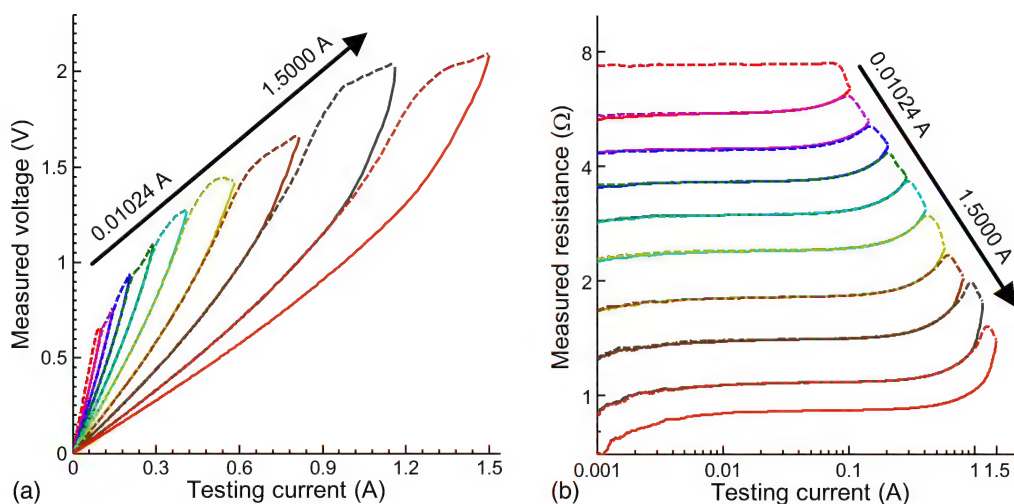


Fig. 5. Typical measured results for a stack of polished samples with nine successive back-and-forth sweeping current cycles with range increasing by a factor of $\sqrt{2}$ from 0.1024 to 1.5000 A (0.1024, 0.1448, 0.2048, 0.2896, 0.4096, 0.5793, 0.8192, 1.1585 and 1.5000 A) under a constant stress of 0.02 MPa: (a) hysteresis loops of voltage; (b) resistance-current characteristics

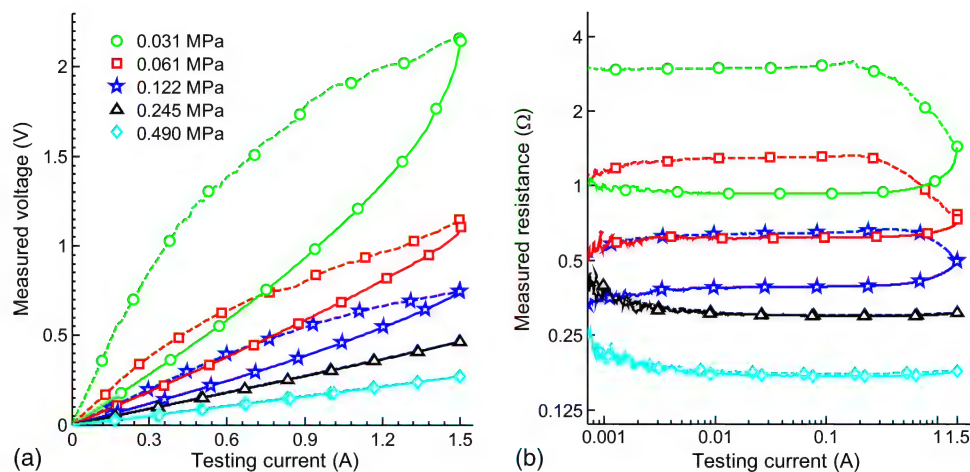


Fig. 6. Typical measured results for polished samples using current sweep under various stresses with solid lines expressing the first phase and the dashed line showing the second phase: (a) variation of the measured voltage; (b) resistance-current characteristics

The adjacent asperities may move closer to each other under a higher contact pressure, which results in the rise of the contacting area, thus enhances the conduction. The enlarged contacting area can carry more current without the microsoldering of the contact points. In engineering practice, the contact resistance is usually reduced by squeezing the contacting components together with greater force, which can bring about a larger true contact area. With the sweep tests shown here, the electrical current seems to be also capable of broadening the current path.

Stress-Dependent Electrical Contact Resistance

The results shown in Figs. 3–6 indicate a likely change of the contact network between the surfaces, where the contacting asperities can be regarded as a network of resistors and capacitors changing with applied current, mechanical load, and time. The true contact area increases linearly with the applied compression, resulting in improved conduction (Kogut 2005; Kogut and Komvopoulos 2005). Meanwhile, the electrical current results in the physical and chemical modification of sample surfaces, which involves many processes, including the rupture of the oxide layer due to compression and the localized heating induced by current.

With the tests presented above, the conclusions are (1) for the studied systems, when the imposed current is less than 50 mA, impact from the current on the measured resistance is negligible, even at low levels of applied stress (lower than 0.020 MPa). However, a small testing current of less than 1 mA can lead to a high-level of measurement noise; and (2) when the conduction time of electrical current (less than 50 mA) is less than 1 s, the influence of the current on resistance can be ignored, especially in cases of high pressures (higher than 0.5 MPa).

For samples exhibiting different surface morphologies, the electrical resistances were measured under various stresses, as shown in Fig. 7. For each type of sample, five series of tests were conducted and the resistances were evaluated at 16 different stress levels from 0.020 to 8.936 MPa. The testing current was set as 10 mA and the measured time was 0.01 s for each individual data point in order to minimize the influence of the testing current on the measurement.

As shown in Fig. 7, the measured resistances of disk stacks with different surface features decrease considerably as the compressive pressure is increased, converging to a value close to the bulk resistance of the material. For the identical loading stress level, samples blasted with 50 μm sized glass beads usually present the highest

resistance among all three types of samples. At low levels of applied stress (less than 0.5 MPa), the measured resistance is spread across a wider range. A few groups of the testing results were not included when jumps occur in the resistance measurement under constant or smoothly increasing loads. The unexpected vibration, electrical noise and some limitations of the experimental setting can contribute to the measurement uncertainty. By fitting the resistance/pressure curves from 0.031 to 1.176 MPa it is observed that the measured contact resistance is a power function of the compressive stress at certain range of loading with the exponents being -0.816 , -1.026 , and -1.494 , respectively, for polished surface (S1), blasted using 300 μm (S2), and 50 μm (S3) diameter glass beads. The absolute values of power exponents demonstrate an increasing trend with the rise of fractal dimension indicated in Table 1. When a loading force of 4,384 N is applied, corresponding to a stress level of 8.936 MPa, the measured resistance can be as small

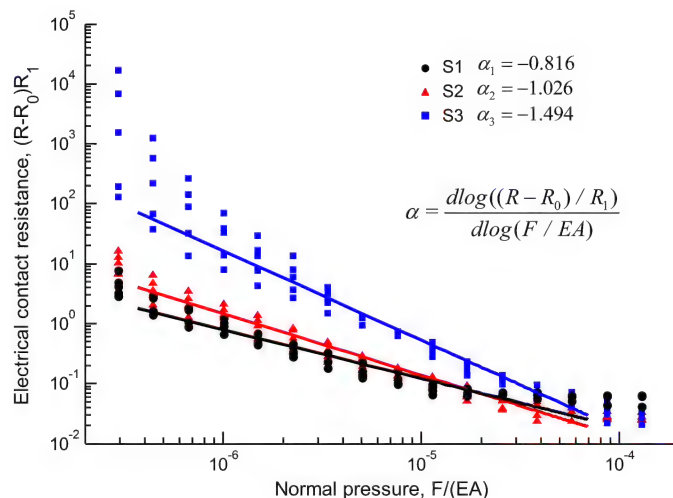


Fig. 7. Stress-dependent ECR of different surfaces under various loading levels with a testing current of 10 mA, with E being the value of the Young's modulus of the tested material, A being the projected area of the tested samples, R_0 being 0.06 Ω , including the combined resistance of bulk material of identical size as the disk stack (equaling 2.53 $\mu\Omega$), wires and connections used in the experimental setting, and R_1 being 1 Ω

as 0.0578, 0.0271, and 0.0303 Ω , corresponding to S1, S2, and S3. The resistance measured at high loads for the three types of tested surfaces appears to be inversely correlated with the values of RMS roughness and RMS slope. Further experiments with more types of surface are necessary to reach a general conclusion on the relationship between the stress-dependent electrical resistance and RMS roughness and RMS slope.

Conclusions

An experimental investigation was performed on the electrical contact resistance of rough surfaces. The experimental results show that the measured contact resistances of a stack of rough aluminium samples rely strongly on surface topology, mechanical loading, as well as testing current and time. The typical resistance creep curves over 200 s were recorded to investigate the current effect on the measured resistance over time. Tests with sweeping current under various compressive loads were carried out to study the influence of the current on the electrical contact resistance. For low-level electrical currents of the orders of a few mA, the electrical resistance is constant, exhibiting a linear ohmic behavior whereas the rise of the electrical current brings about a decrease of the electrical resistance. An increased compressive load results in the weakening of the effects of testing current on the measured resistance and decreases the time dependent resistance relaxation. Similarly, a higher testing current can also cause the reduction of the impact from the loading force. With testing current set as 10 mA and the measuring time being 0.01 s for each measured resistance, the impact of the current can be ignored for all three types of samples under varying loading levels. Under this condition, the measured resistances of stacks of samples decrease continuously under increasing stresses, approaching the resistance for bulk material at high loads. The results also demonstrate that a power law relationship exists between the measured electrical resistance and normal stress across a certain stress range, with the absolute values of exponents rising with the fractal dimension of the surfaces.

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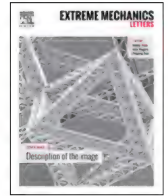
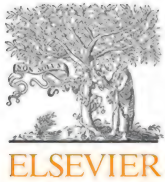
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4.2 Paper 4: Interfacial electro-mechanical behaviour

The inter-particle electrical property is playing a significant role in governing the electrical transport of granular materials. This paper presented here studies interfacial stiffness and electrical contact resistance, which are both closely related to real contact area, allow us to understand how two contacting solids with rough surfaces interact with each other over a wide range of length scales. The results of our original experiments show that the interfacial contact stiffness and electrical conductance follow power-law functions with respect to the normal force over a wide range of applied loads. However, different values of power-law exponents were found. Specifically, the power-law exponent for electrical contact resistance is approximately double of that for normal interfacial stiffness. This is because the transport behaviour at the rough contact at macro scales is dominated by the contact behaviour at lower scales. Moreover, the microstructures represented by surface roughness bring about strong interconnections of electrical-mechanical coupling. This work provides a first-order investigation connecting interfacial mechanical and electrical behaviour, applicable to studies of electrical transport in granular materials with numerous contacts. Both the interfacial stiffness presented in Chapter 3 and electrical contact resistance shown in this chapter are fundamentally determined by the real contact area at rough interface. By relating the two parameters, one can obtain a comprehensive understanding of the rough contact and the electrical transport at rough interfaces. The information obtained in Chapter 3 and 4 will be scaled upwards to granular systems at macroscale through contact network built by individual inter-particle contacts to interpret the stress-dependent electrical conduction in granular materials, shown in the following two chapters.

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Interfacial electro-mechanical behaviour at rough surfaces



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ABSTRACT

In a range of energy systems, interfacial characteristics at the finest length scales strongly impact overall system performance, including cycle life, electrical power loss, and storage capacity. In this letter, we experimentally investigate the influence of surface topology on interfacial electro-mechanical properties, including contact stiffness and electrical conductance at rough surfaces under varying compressive stresses. We consider different rough surfaces modified through polishing and/or sand blasting. The measured normal contact stiffness, obtained through nanoindentation employing a partial unloading method, is shown to exhibit power law scaling with normal pressure, with the exponent of this relationship closely correlated to the fractal dimension of the surfaces. The electrical contact resistance at interfaces, measured using a controlled current method, revealed that the measured resistance is affected by testing current, mechanical loading, and surface topology. At a constant applied current, the electrical resistance as a function of applied normal stress is found to follow a power law within a certain range, the exponent of which is closely linked to surface topology. The correlation between stress-dependent electrical contact and normal contact stiffness is discussed based on simple scaling arguments. This study provides a first-order investigation connecting interfacial mechanical and electrical behaviour, applicable to studies of multiple components in energy systems.

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1. Introduction

Interfacial electro-mechanical behaviour is fundamental to indicators of energy system performance such as electrical power loss, cycle life, and storage capacity in lithium-ion batteries [1,2], sodium-ion batteries [3], solid oxide fuel cells [4], photovoltaics [5] and thermoelectric systems [6]. Surface morphology plays an essential role in determining how contacting solids interact with one another in a variety of processes including thermal swelling, electrical conduction, electrochemical reactions, friction,

and adhesion [7–9]. In energy storage and conversion applications the effective mechanical and electrical properties of granular electrode structures can be connected to microstructural characteristics [2,10]. However, the interfacial properties in the existing modelling approaches are usually simplified [11].

Energy losses at interfaces are usually associated with ohmic heating (also known as Joule heating) due to the passage of an electrical current through contacting surfaces. In the context of energy management, improved electrical contacts play a prominent role in mitigating energy losses in battery assemblies. The energy loss due to the electrical contact resistance (ECR) at interfaces between electrode layers and at contacts between electrodes and current-collectors can be as high as 20% of the total energy flow of the batteries under normal operating conditions [12,13].

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The effects of the mechanical properties and surface roughness of electrical contacts on the performance of electrical connectors are of great importance in terms of potential drop and heat accumulation in contact zones [14,15]. A significant increase in ECR can be caused by interfacial resistance due to the inevitable presence of resistive surface films, including corrosion deposits, fracture debris, oxide and hydrated layers at electrical contacts, resulting in excessive ohmic heating. In extreme cases, the heat can bring about system failure through sparks, fire and even melting of system components [12,16,17].

The stress dependence of ECR at rough surfaces can be associated with the varying true interfacial contact area during system operation. However, the direct quantitative evaluation of real interfacial contact area between bodies through either experimental measurements or numerical simulations remains highly challenging due to the complex multi-scale morphologies exhibited by rough surface structures [18–20]. Significant difficulties remain in relating interfacial electro-mechanical properties to surface structure descriptors.

Employing electrical measurement, nanoscale mechanical testing and surface morphology characterisation, we investigated interfacial normal contact stiffness and electrical conduction behaviour at rough interfaces with random multiscale morphologies. First, we conducted contact stiffness measurements using flat-tipped diamond nanoindentation tests on a set of rough surfaces. Then, we examined the evolution of electrical conduction with varying compressive loads. Based on these results, discussions are extended to the relationship between electrical contact conductance and contact stiffness. This study demonstrates the importance of a multi-physics understanding of the origins of the electro-mechanical behaviour at interfaces in order to improve the reliability and performance of electrical contacts in energy systems.

2. Theoretical background

Compared with the apparent or nominal contact area, the true area of contact at an interface is considerably smaller due to the existence of surface roughness. As shown in Fig. 1, when an electric current is conducted between two contacting solids, the restricted contact area, which depends on the size and spatial distribution of contacting asperities, causes additional constriction resistance (known as the electrical contact resistance, ECR) [21]. In addition to the constriction resistance resulting from the limited areas of true contact at an interface, ECR is also affected by the existence of resistive surface films, such as oxide layers [8]. Theories of ECR have since been further developed to include the effects of elastic–plastic deformation of the contacting asperities due to applied forces, multi-scale surface topography, size effects, and the contribution of insulating films between contacting bodies [18,22–25].

Current flowing through rough interfaces is scattered across a large number of micro-contacts of various geometries, which are often assumed to be circular in theoretical treatments [19,26]. The constriction resistance due to the convergence and divergence of current flow

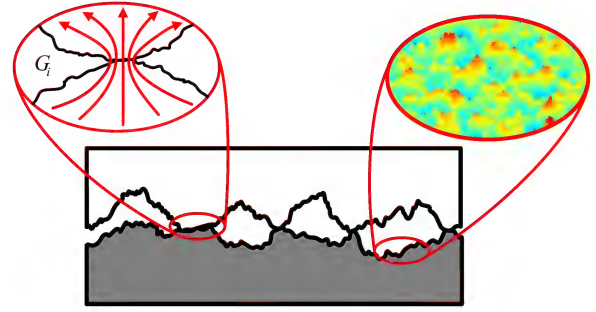


Fig. 1. Schematics of electrical conduction through a rough interface exhibiting multi-scale surface features.

at a single contact is represented in Fig. 1. The resistance of a single contact is dictated by the dominant electronic transport mechanism, which depends on the contact area and structure. When the radius of the micro-contact, r , is comparable or smaller than the average electron mean free path, λ , the constriction resistance is dominated by the Sharvin mechanism, in which electrons travel ballistically across the micro-contacts. The resistance of a contact with area, a , is given by [27].

$$R_S = \frac{\lambda(\rho_1 + \rho_2)}{2a} \quad (1)$$

where ρ_1 and ρ_2 are the specific resistivities of the contacting surfaces. On the other hand, when $r > \lambda$, the electron transport through the contact can be treated classically (Holm contact). The resistance can be expressed in the following form [8]:

$$R_H = \frac{\sqrt{\pi}(\rho_1 + \rho_2)}{4\sqrt{a}}. \quad (2)$$

The total electrical conductivity, G_c , of an interface is assumed to be the sum of individual conductivity $G_i = R_i^{-1}$ at micro-contacts, corresponding to the restriction resistances in parallel:

$$G_c = \sum G_i. \quad (3)$$

In the case of rough surfaces with multiple contacting asperities, there is a distribution in the size of the contact area. The Sharvin and Holm expressions should therefore be considered as limiting cases.

In general, the area of contacts used in Eqs. (1) and (2), and therefore the contact resistance, depends on the applied pressure. Using theoretical and numerical approaches [18,22,28,29], power-law type semi-empirical correlations between the contact resistance and the normal pressure have been proposed for rough interfaces. In particular, previous theoretical studies found the contact conductance to be linearly proportional to the incremental stiffness [18,22].

It should be noted that many mechanisms of surface structure evolution have been observed during electrical conduction through rough interfaces, including dielectric breakdown of oxide layers, localised current-induced welding, chemical disorder arising with random composition and oxidation processes in corrosive environments, and surface diffusion [30,31]. These phenomena are outside the scope of this paper.

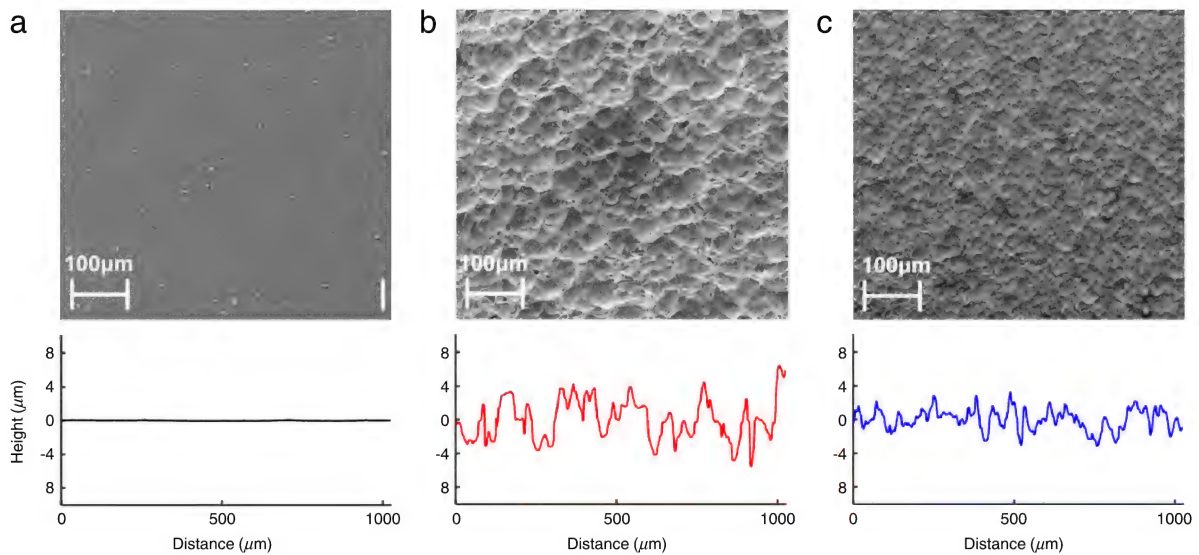


Fig. 2. SEM images and typical surface profiles of aluminium samples subjected to different surface treatments: (a) polished, S1; (b) sand blasted with 300 μm -sized glass beads, S2; (c) sand blasted with 50 μm -sized glass beads, S3.

Table 1
Sample surface characteristics for different surface treatments.

Sample type	RMS roughness/ μm	Fractal dimension, D_f	RMS slope	Contact stiffness, α_E	Electrical conductance, α_G	Exponent ratio, α_G/α_E
S1	0.057 ± 0.005	2.093 ± 0.062	0.009 ± 0.001	0.463 ± 0.022	0.816 ± 0.081	1.762 ± 0.194
S2	4.179 ± 0.194	2.551 ± 0.022	0.224 ± 0.015	0.569 ± 0.029	1.026 ± 0.049	1.803 ± 0.126
S3	2.970 ± 0.276	2.626 ± 0.017	0.202 ± 0.010	0.605 ± 0.022	1.494 ± 0.134	2.469 ± 0.239

3. Surface preparation and characterisation

Round disks, with a diameter of 25 mm, made of aluminium alloy 5005 were used to fabricate specimens for both the measurement of the interfacial contact stiffness and ECR. For each individual sample, both the top and bottom surfaces were subjected to the same treatment using standard polishing and sand blasting procedures. The average diameters of the two selected groups of glass beads used in blasting treatments were 50 μm and 300 μm . The sand blasting process was conducted for one minute, a duration which was sufficient to yield homogeneously and isotropically modified surface features. The sample surfaces were fabricated in such a way that each set of surfaces exhibited a distinct combination of surface roughness indicators, namely root mean squared (RMS) roughness and fractal dimension. Fig. 2 shows scanning electron microscope (SEM) images and typical surface profiles of the different surface types used in this work. Based on the three-dimensional digitised topographies obtained by optical surface profilometry (NanoMap 1000WLI), the mean values of RMS roughness, fractal dimension and RMS slope with standard deviations over ten scans on different samples were calculated, as shown in Table 1. These values are found to be comparable with descriptors of naturally occurring surfaces [32]. Values of RMS roughness were calculated as the RMS average of the profile height from the scanning. In the digitised scanning, the slopes of triangular units formed by three adjacent pixels are used to calculate

the RMS slope, which is commonly chosen as a higher order surface descriptor [33,34]. The scaled triangulation method [34] was used for the calculation of fractal dimension values. It was found that the smaller the particles used to modify the surfaces the larger the fractal dimension was. The fractal dimension, a cross-scale surface descriptor that incorporates localised and macroscopic surface information provides an effective means for modelling engineering surfaces with random self-affine multi-scale properties in the characterisation of surfaces and particles [35]. The advantage of using surface fractality as a cross-scale surface descriptor stems in part from the tendency of first order descriptors (e.g., maximum height or mean roughness of the surface) to be dominated by highest scale features, while secondary descriptors (e.g., slope) tend to be dominated by finest scale surface characteristics [33,34].

4. Contact stiffness at rough surfaces

The surface contact stiffness of aluminium samples with different surface morphology was assessed using nanoindentation (Agilent G200) with three flat indenter tips of different diameters of 54.1 μm , 108.7 μm , and 502.6 μm (FLT-D050, FLT-D100, and FLT-D500, respectively, SYNTON-MDP, Switzerland). The reason for choosing flat tips is that the apparent contact area under the tip does not change with respect to the indentation depth, which is not the case for spherical or Berkovich tips. When the flat indenter tip first comes into contact

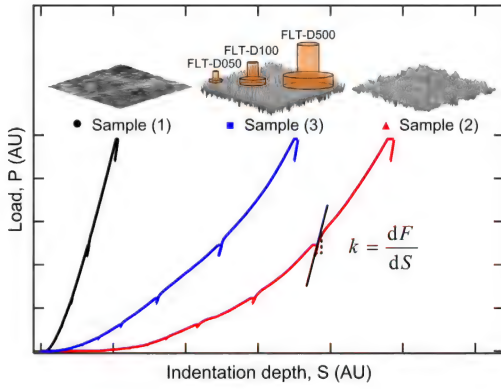


Fig. 3. Typical loading-displacement curves of nanoindentation tests on three types of surfaces. Ten partial unloading tests were carried out to isolate elastic contributions to contact stiffness under different loading levels. The three flat indenter tips used in the experiments (FLT-D050, FLT-D100, FLT-D500) are also illustrated for comparison.

with the testing sample, the actual contact area is only a small fraction of the nominal contact area. The asperities of the sample surface at contact regions are then squeezed against the flat tip as indentation progresses as is shown in Fig. 3. In order to evaluate only the elastic responses, partial unloading tests were successively performed at ten intervals by decreasing the applied load by 10% each time. The loading level of each subsequent unloading stage is twice that of the previous unloading stage, with a maximum load of 500 mN during the last unloading step.

Mean stiffness values were obtained by averaging data of ten indentation tests at different locations for each surface type. The unloading stiffness is here defined as the initial slope of the unloading curve, $k = dF/dS$, where F designates the normal force and S is the indentation depth. Subsequently, the reduced elastic modulus E_r was derived from the measured unloading stiffness as

$$k = \beta \frac{2}{\sqrt{\pi}} E_r \sqrt{A}, \quad (4)$$

where A is the apparent contact area of the indenter tip and β is a geometrical constant, taken as unity for a flat punch [36]. Eq. (4) is a fundamental equation for assessing the elastic properties in nanoindentation tests. The reduced modulus depends on the elastic properties of both the tested specimen and the indenter tip:

$$\frac{1}{E_r} = \frac{1 - \nu_c^2}{E_c} + \frac{1 - \nu_i^2}{E_i}, \quad (5)$$

where ν_i and ν_c represent the Poisson's ratios of the indenter tip material and the tested specimen respectively. For the diamond indenter tips used in this research, E_i and ν_i are typically 1140 GPa and 0.07, respectively. Eqs. (4) and (5) allow the estimation of E_c from measured values of A and k , while for ν_c we simply use the Poisson's ratio ν^* of bulk aluminium ($\nu_c = \nu^* = 0.3$).

By using different sized flat tips, the stress range extends over several orders of magnitude. With the same maximum force (500 mN) provided by the nanoindenter, the maximum stress produced with FLT-D050 was around 100 times larger than that produced with FLT-D500. The

stress provided by all the three indenter tips ranged from 0.005 MPa to 200 MPa, spanning five orders of magnitude. The contact stiffness measured over this range of applied stresses varies approximately from 0.01 GPa to 55 GPa.

Fig. 4 shows the evolution of contact stiffness with applied force for the different surfaces. Here, we normalised the contact elastic modulus E_c by the Young's modulus of aluminium alloy 5005, $E^* = 69.5$ GPa. The force is normalised by E^*A , where A is the projected area of the corresponding tip. The measured contact stiffness increases with the loading force, for all tested samples. At the same applied stress level, the surfaces after sand blasting treatment (samples 2 and 3) show a smaller value of contact stiffness with respect to that of the polished surface (sample 1). The surface blasted with glass beads of 50 μm diameter (sample 3) presents the lowest contact stiffness of all the three types of surfaces.

We express here the power-law relation of the contact stiffness with the applied normal force

$$E_c \propto (F)^{\alpha_E}, \quad (6)$$

where α_E is the exponent of the power-law function [37,38]. It should be noted that the fitting curves are achieved excluding the contributions from the measured stiffness under stress levels higher than 100 MPa, where the surface shows an apparent yield phenomenon. For all the three surface types, the value of the exponent α_E varies from 0.4626 to 0.6048 (in Table 1), changing as the fractal dimension increases. In comparison, the typical value in cases of Hertzian contact of two elastic spheres is 1/3, as shown in Section 6. The power-law relationship found here experimentally is in good agreement with previous theoretical predictions on a quantitative basis [18,37,38].

5. Electrical conductance at rough surfaces

For each surface type, interfacial electrical conductance was measured for stacks of eleven disks, giving ten rough-to-rough interfaces. Analysis was achieved by means of a source/measurement unit (SMU B2900A, Agilent) across a range of applied compressive loading forces. In this experimental setting, we measured the resistance created by ten interfaces instead of a single interface, aiming to achieve a higher precision, larger linear range and better robustness against the measurement noises from the connecting wires, loading device and measurement unit. Using multiple interfaces further reduces experimental errors arising from inhomogeneity in surface treatment processes.

Prior to the measurement of force-dependent resistance, we performed resistance creep tests and sweeping current tests to select the most appropriate testing current and time to minimise influences on the measurement from the applied current. Full procedures and results have been previously published in greater detail [17]. The applied sweep current test consisted of two phases: a "loading" phase (P1) with current increasing logarithmically from 0.0001 A to 1.5 A, followed by an "unloading" phase (P2) with current decreasing logarithmically from 1.5 A back to 0.0001 A. Both phases were conducted under conditions of

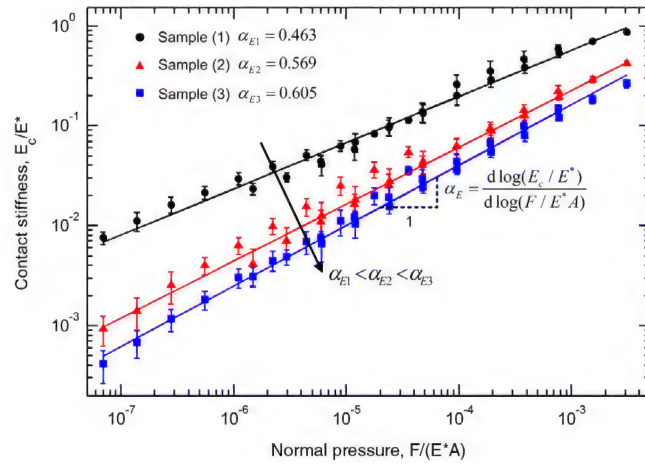


Fig. 4. Curve fitting for the normalised stiffness, E_c/E^* , and the normalised applied force, $F/(E^*A)$, for three tested surfaces, with E^* being the Young's modulus of the tested material, and A the apparent contact area.

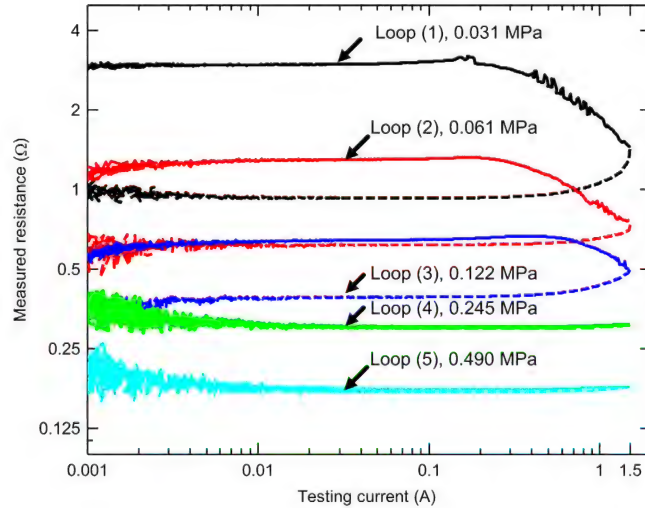


Fig. 5. Typical measured results for polished samples using current sweep under various stresses (0.031 MPa, 0.061 MPa, 0.122 MPa, 0.245 MPa and 0.490 MPa, corresponding to loops 1–5, respectively) with solid lines representing the first phase (P1) and the dashed lines showing the second phase (P2).

constant normal load. During the sweeping loops, the voltage was recorded at a frequency of 2 kHz. The two-phase sweeping process was completed within 0.2 s in order to avoid significant time dependent resistance degradation.

Fig. 5 shows the typical resistance–current characteristics for polished samples obtained from sweeping current tests. Each individual loop corresponds to a distinct load. The five loops shown demonstrate similar trends known as the Branly effect [30,31], i.e., the measured resistance begins to drop irreversibly after the testing current reaches a certain value. The process is featured by voltage creep, hysteresis loops, and voltage saturation effects [31,39]. The corresponding threshold current values for loops (1–3) are approximately 150 mA, 200 mA and 400 mA, respectively, and the value seems to be positively correlated with the applied normal load. However, the Branly effect tends to be harder to capture at sufficiently high stress levels, shown in loops (4–5). For all five loops, when the testing current is higher than approximately 5 mA and lower than

the threshold current values, the measured resistances remain stable at two plateaus in both P1 and P2, and can therefore be defined as ohmic resistance (the testing current is directly proportional to the measured voltage). At low testing currents (lower than 1 mA), the measured resistance exhibits instability with the prominent measurement noises. The measured resistance obtained from subsequent sweeping tests will follow the path of the unloading phase (shown in the dashed lines in Fig. 5) [17].

The experimental results in Fig. 5 indicate that both mechanical loading and electrical current alter the surface morphology and broaden the gap in measured resistance between P1 and P2. The contacting asperities can be regarded as a resistor network changing with the applied current, mechanical load, and measurement time. At the interfaces, the electrical current results in the physical and chemical modification of sample surfaces, which involves many processes, including the rupture of the oxide layer due to compression, and the localised heating induced

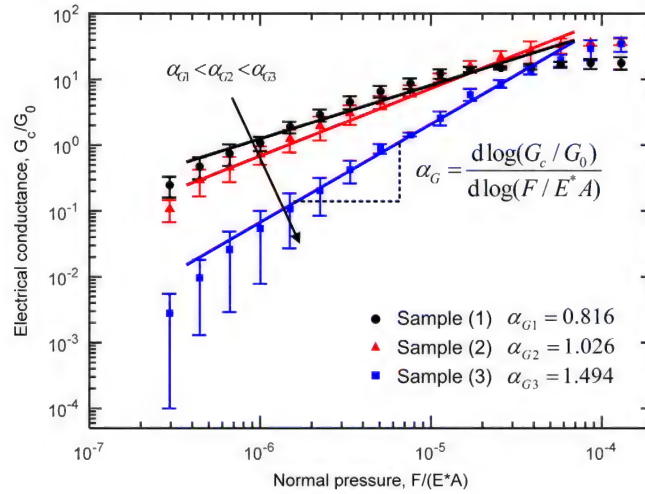


Fig. 6. Stress-dependent electrical conductance of different surfaces under various loading levels with a testing current of 10 mA, with E^* being the value of the Young's Modulus of the tested material, A the projected area of the tested samples, and G_0 being set to $1 \text{ } \Omega^{-1}$. The curve fitting were conducted for loading levels in a range of [0.031 MPa, 3.973 MPa].

by current. A high level of applied stress leads to better stability and repeatability of ECR measurements [17,31].

Based on the performed sweeping current tests the electrical resistance was measured under various stresses, for samples exhibiting different surface morphologies. For each type of sample, five series of tests were conducted and the resistances were evaluated at 16 different stress levels from 0.020 MPa to 8.936 MPa. The measured time was 0.01 s for each individual data point, in order to avoid significant effects arising from ohmic heating and associated time dependent resistance degradation. The testing current was set at 10 mA, where all the three types of samples display an ohmic behaviour under varying electrical and mechanical loads. The interfacial electrical contact conductance was subsequently calculated through $G_c = 1/(R_c - R_0)$, where R_0 ($\sim 0.06 \text{ } \Omega$) is the combined resistance of the bulk material of identical size as the disk stack ($\sim 2.53 \text{ } \mu\Omega$), wires and connections used in the experimental setting.

As shown in Fig. 6, the measured conductance of disk stacks increases considerably with pressure, converging to a value close to the bulk conductance of the material. For given stress levels (≤ 4 MPa), samples blasted with $50 \text{ } \mu\text{m}$ sized glass beads (S3) usually present the lowest conductance among all the three types of samples. At low levels of applied stress (less than 0.5 MPa), the conductance is spread across a wider range. Similar to Eq. (6), we use a power-law function to express the correlation of the conductance with applied normal load as

$$G_c \propto (F)^{\alpha_G} \quad (7)$$

By fitting the conductance/pressure curves from 0.031 MPa to 3.973 MPa, the power law exponent α_G is found to be 0.816, 1.026 and 1.494 respectively for polished surfaces (S1), surface blasted with $300 \text{ } \mu\text{m}$ particles (S2) and those treated with $50 \text{ } \mu\text{m}$ particles (S3). The exponent values increase with the fractal dimension, shown in Table 1. Moreover, for all the three types of surfaces as shown in Fig. 6, the electrical conductance reaches a plateau

under higher stresses, with the plateau value correlating to the RMS roughness. In the lower stress regime, the experimental data no longer seems to follow the power law.

6. Discussion

The key experimental results for contact stiffness and electrical conductance, measured for three types of rough surfaces, are summarised in Table 1. Both the contact stiffness and electrical conductance increase with the applied force, exhibiting power law behaviours with exponents α_E and α_G , respectively. These exponents vary with the surface roughness and increase with the fractal dimension. In contrast, no evident correlation between the RMS roughness value and the exponent was found. This suggests that the correlation between contact stiffness, electrical conductance and applied force is dominated by fine scale surface characteristics.

We rationalise the experimental findings by developing the following scaling arguments. Both the contact stiffness and conductance primarily depend on the true contact area A_c , which evolves during mechanical loading and cannot be determined in a direct way based on the considered measurement methods. As a workaround, we estimate the true contact area based on the following expression for the incremental stiffness:

$$k = \beta' \frac{2}{\sqrt{\pi}} E_r' \sqrt{A_c}, \quad (8)$$

where E_r' is the (constant) reduced elastic modulus calculated for the bulk elastic properties of the tested material and indenter: $E_r' = ((1 - \nu^{*2})/E^* + (1 - \nu_i^2)/E_i)^{-1}$, and β' is a geometric factor of the order of unity. By writing Eq. (8), we assume that the effect of surface roughness on the measured incremental stiffness can be described by considering the true contact area A_c (rather than the project contact area A) and bulk material properties in the

fundamental Eq. (4). By comparing Eq. (4) and (8), and considering that $E_i \gg E^* > E_c$, we obtain the following scaling relation:

$$E_c/E^* \propto \beta' \sqrt{A_c/A}. \quad (9)$$

Next, we consider the true contact area to be the sum of n individual contact areas, with average $a = A_c/n$. Here, individual asperities are assumed not to interact with one another during deformation. In order to relate the evolution of a to the applied force, we first rely on a classical result of Hertzian contact theory. Representing a single contact by two spheres with radii R_1 and R_2 squeezed against each other, the contact area varies with the applied force according to

$$a = \pi \left(\frac{3RF}{4E'_r} \right)^{2/3}, \quad (10)$$

where $R = (1/R_1 + 1/R_2)^{-1}$ is the equivalent radius of the two spheres, and the reduced modulus E'_r was introduced in Eq. (8). Eqs. (9) and (10) indicate that the contact stiffness is a power function of the load, with an exponent 1/3. This simple scaling analysis is not consistent with our experimental findings for α_E , which takes significantly higher values. However, the scaling analysis based on the Hertzian contact theory does not consider the changing number of contact asperities, n , for the increasing load. Furthermore, at the rough interface, the contact areas are not uniformly distributed [40], and interactions between asperities can exhibit complex deformation mechanisms, such as plastic deformation, adhesion, and friction.

On the other hand, introducing relation (10) into Eqs. (1) and (2) for the Holm and Sharvin resistance at a single contact, one finds that:

$$G_H = \frac{4}{\tilde{\rho}} \left(\frac{3RF}{4E'_r} \right)^{1/3}, \quad G_S = \frac{2\pi}{\lambda \tilde{\rho}} \left(\frac{3RF}{4E'_r} \right)^{2/3}, \quad (11)$$

where $\tilde{\rho} = (\rho_1 + \rho_2)$. Combining (11) with (3), we find that the total conductance of the rough surface, G_c , approximately scales with the force following a power law with the exponent ranging from 1/3 (Holm) to 2/3 (Sharvin), depending on the dominant conduction mechanisms at individual contacts.

We further consider the contact model for a conical punch [41], where the contact area, a , is found to be linearly proportional to the applied force, F . With the same analysis as above, an exponent $\alpha_E = 0.5$ can be derived for the contact stiffness, and an exponent $\alpha_G = 0.5 \sim 1$ for the electrical conductance. This provides a better representation for the exponents of contact stiffness and electrical conductance as compared to the prediction by the Hertzian solution. Again, the exponents derived from this simple scaling analysis are lower than the experimental values of α_G . This may also be due to the fact that the scaling neglects the increase in number of contacting points under increasing compression.

Despite these discrepancies, it is interesting to consider the ratio of the exponents for contact stiffness and electrical conductance, α_G/α_E . This ratio characterises the power law relation between the conductance and contact

stiffness, with $G_c \propto (E_c)^{\alpha_G/\alpha_E}$. According to the scaling analysis, this ratio ranges from 1 (Holm mechanism) to 2 (Sharvin mechanism). Experimentally, an approximate value of 2 was found for loading levels in a range of $F/(E^*A) \in [5 \times 10^{-7}, 5 \times 10^{-5}]$. For sample 3, similar fitting in the low load region gives a higher value of α_G , and hence a higher value of the ratio α_G/α_E , suggesting a larger proportion of Sharvin-type contacts at low loads. Similar observations seem to hold as well for samples 1 and 2, but the transition takes place at even lower loads. As the load increases, new asperities come into contact, the contacting points enlarge and small microcontacts merge forming large contacts, resulting in better conduction. The dominant conduction mechanism transitions from a Sharvin-type to a Holm-type with the exponent ratio decreasing from 2 to 1. Under sufficiently high forces, and hence high contact areas, the electric and mechanical properties converge to those of the bulk material, as expected.

The ratio α_G/α_E also tends to increase with the fractal dimension. A surface with a higher fractal dimension demonstrates Sharvin dominated conductance ($\alpha_G/\alpha_E \sim 2$), while a less fractal surface presents combined Sharvin and Holm-type conductance (α_G/α_E between 1 and 2).

Note that in the contact stiffness measurements, the flat indenter tips can be considered as rigid flat surfaces ($E_i \gg E^*$), corresponding to a rough-to-flat contact problem. In comparison, our interfacial electrical resistance experiments involve rough-to-rough contacts. However, a scaling analysis based on rough-to-flat contact would yield identical exponents in the power law functions (10) and (11) [28,33].

In the experiments, both contact stiffness and conductance may be affected by oxide layers at the sample surfaces. Aluminium alloys ubiquitously exhibit thin passivated hydrous and oxide layers arising from reaction with atmospheric oxygen and water. This nanoscale layer exhibits locally divergent mechanical properties in a region of thickness typically less than 10 nm, which is significantly less than the depth of indentation performed in the current work. The influence of oxide layers is thus expected to be of limited significance in the present contact mechanics study. In the analysis of ECR behaviour, the oxide layer acts as an insulator. However, due to its limited thickness, the measured conductance is only sensitive to the presence of this layer at lower loads. For this reason large measurement uncertainties are evident at low loads with the magnitude of these fluctuations dependent on specimen surface structure, as shown in Fig. 6. Therefore in this study the effect of the oxide layer is minimal and does not interfere with the findings.

The observations made in this study can provide insights into the physical origin of the topological dependence of transport phenomena in energy materials applied in conversion, storage and generation systems. Parametric studies in to the performance of energy systems often yield unexpected behaviour arising from changes to the structure or processing of complex materials such as granular electrodes [2,10]. The present work suggests that the structure and mechanics of interfaces in these systems may be in part a contributing factor to the observed processing dependence of performance.

7. Conclusion

We performed experimental investigations into the contact stiffness and electrical contact resistance at rough interfaces, with a specific focus on their dependence on applied force. The change of these interfacial electro-mechanical properties under different loading conditions can be associated with changes in the true area of interfacial contact. The measured contact stiffness and electrical conductance have been found to exhibit power law relationships with normal pressure across a wide range of applied stress, expressed as $E_c \propto (F_N)^{\alpha_E}$ and $G_c \propto (F_N)^{\alpha_G}$, respectively. The corresponding exponents of these relationships were found to be closely correlated to surface fractality with the absolute values of α_E and α_G , increasing with the fractal dimension of the surfaces. The presented experiments on load-dependent contact stiffness and electrical contact resistance provide an initial step towards connecting interfacial electro-mechanical properties and surface topology, which is of value in interpreting the properties of various energy materials and components. Further investigation is warranted to fully understand these phenomena and interpret the interface-morphological dependence of energy material performance.

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5 Electrical Transport in Binary Percolation Networks

Granular materials are actually multi-contact systems with particles. The electrical transport in many types of granular materials is realized by current paths formed by the inter-particle contact, the electrical properties of which depends on the inter-particle force and the interface structure, as shown in Chapter 3 and 4. The overall conduction behaviour can be simulated by resistor-capacitor networks. Specifically, each individual network node is a particle of the considered granular packing, and each individual network element, such as a capacitor or a resistor, represents a contact in the granular materials. In this chapter containing Paper 5, we studied the influences of network geometry and boundary conditions on the electrical responses of random binary percolation networks. For varying network configurations in terms of network aspect ratio and electrode dimension, a unified description focusing on the centre and the span of the emergent scaling region is provided for the universal dielectric responses. The findings of this paper can guide the design and testing of finite disordered systems with multi-contacts.

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RESEARCH ARTICLE

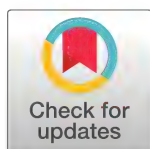
Universality of the emergent scaling in finite random binary percolation networks

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Abstract

In this paper we apply lattice models of finite binary percolation networks to examine the effects of network configuration on macroscopic network responses. We consider both square and rectangular lattice structures in which bonds between nodes are randomly assigned to be either resistors or capacitors. Results show that for given network geometries, the overall normalised frequency-dependent electrical conductivities for different capacitor proportions are found to converge at a characteristic frequency. Networks with sufficiently large size tend to share the same convergence point uninfluenced by the boundary and electrode conditions, can be then regarded as homogeneous media. For these networks, the span of the emergent scaling region is found to be primarily determined by the smaller network dimension (width or length). This study identifies the applicability of power-law scaling in random two phase systems of different topological configurations. This understanding has implications in the design and testing of disordered systems in diverse applications.

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Introduction

The bulk behavior of complex systems comprising disordered multi-phase components is of importance in diverse applications including supercapacitors and batteries [1–4], dielectric material characterization [5–10], the mechanics of structures [11–13], fracturing process [14], thermal analysis [15] and soil probing [16]. In such systems, various parameters govern the electrical, thermal, chemical, and/or mechanical properties of components of a system across multiple scales from molecular up to macroscopic length-scale. Experimental and computational research efforts are increasingly conducted in order to gain insights into the manner in which these properties combine across scales to determine overall system performance.

In particular, the AC conductivity of systems that can be schematically represented as mixtures of electrical components has been the subject of numerous investigations that have shown power-law scaling with frequency arising through different relaxation mechanisms [11, 17–19]. Above a critical frequency this scaling of AC conductivity is described by Jonscher's power law [20] and has been experimentally observed across diverse conductor-dielectric

composites and porous materials [5, 7, 18, 21, 22] with this scaling being termed the “Universal Dielectric Response” [17, 18, 21, 23]. This emergent property does not arise directly from any particular physical or chemical properties of the involved components, but rather is a consequence of the way components combine [11, 19, 21, 24, 25]. Such dielectric mixtures have been effectively approximated as a random network of resistors and capacitors [18, 19, 24] with representative conductors exhibiting a constant conductance $1/R$ and dielectric components exhibiting a variable complex admittance $i\omega C$, which is directly proportional to an angular frequency ω , as illustrated in Fig 1. Useful asymptotic formula for the emergent network admittance including both the effects of component proportions and the network size can be obtained based on the spectral method [21, 26] and the averaging approach [11, 21, 27]. However, establishing a more rigorous estimation necessitates numerical analysis.

From previous numerical studies [11, 17–19, 21, 23, 25, 28], the typical obtained conductivity-frequency spectrum of a square lattice resistor-capacitor (RC) network can be divided into three regions of angular frequency, ω , governed by the proportion of capacitors, p_c , and network size, N (a) an emergent region for intermediate frequencies; (b) two percolation regions; (c) transitions between the two above-mentioned [21]. Symmetry is found between the overall responses at high and low frequencies, which can be correlated to percolation behavior [26, 29, 30]. For low and high frequencies, current tends to percolate predominantly through resistors and capacitors respectively, as these will exhibit relatively lower impedance, as presented in Fig 1. For intermediate frequencies, where the values of admittance for the resistors and the

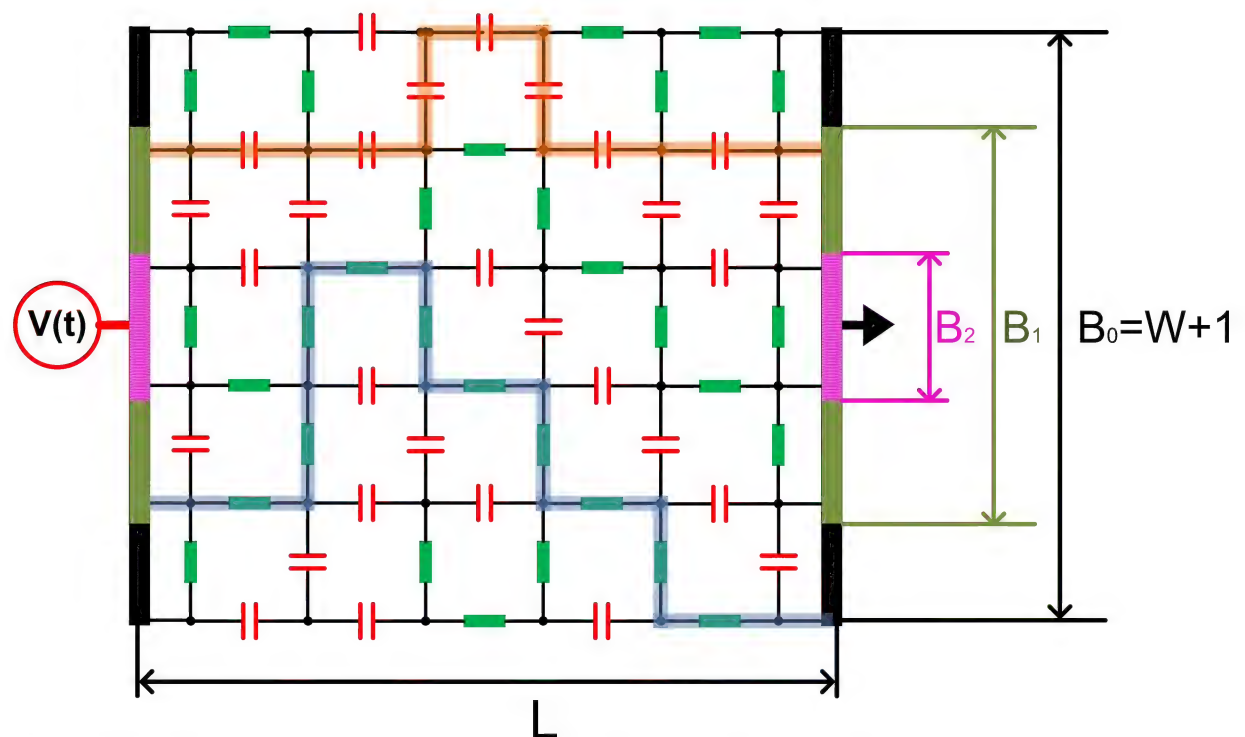


Fig 1. A lattice network containing $W \times L = 5 \times 5$ randomly distributed resistors and capacitors. The network width W and length L indicate the numbers of horizontal and vertical elements in a single chain, respectively. The values of $B_0 = 2$, $B_1 = 4$, and $B_2 = 6$ present the number of nodes connected directly to the electrodes. Percolation paths formed by resistors and capacitors are shown in thick blue and orange lines, corresponding respectively to the dominating modes at low and high frequencies, for the B_0 configuration.

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capacitors are close, we observe the power-law emergent behavior whereby conductivity is proportional to ω^α , with $\alpha \approx p$ [11, 21, 27].

Motivated by the applicability of these networks for representing real-world disordered systems, in this paper we reexamine the universality of the emergent power-law scaling observed in previous work by further considering the significance of the network aspect ratio and boundary conditions on the convergence point and the span of the emergent region, the two key network characteristics of universal scaling behavior.

Methods

In this paper, we extend the square lattice RC networks to rectangular ones with $N = W \times L$ elements distributed between two bus-bars, one of which is grounded and the other raised to a potential, $|V_0|e^{i\omega t}$. A system of complex number linear equations is set up by applying Kirchhoff's current law (for complex currents) on each individual node of the RC network. For the node k , we get

$$I_k(t) = \sum_j^n |I_{j,k}| e^{i(\omega t + \varphi_{j,k})} = \sum_j^n (|V_{j,k}| e^{i(\omega t + \varphi_{j,k})} - |V_k| e^{i(\omega t + \varphi_k)}) / Z_{j,k} = 0, \quad (1)$$

where I_k is the sum of the currents (negative or positive) flowing towards the node k , from connected components. The impedance of a component connected to node k , $Z_{j,k}$, is randomized to be either R or $1/i\omega C$. The voltage potential of the connected node j , $|V_{j,k}| e^{i(\omega t + \varphi_{j,k})}$, with respect to that of the node k , is represented as complex-valued function of time, t , with $\varphi_{j,k}$ being the relative phase. The value of n is determined by the location in the network, equaling the number of connected components. More specifically, $n = 4$ for ones located away from the boundaries, $n = 2$ for the lattice corners, $n = 3$ for the nodes on the boundaries excluding corners. The two electrodes are also regarded as nodes with $n = B$. The electrode dimension, B , is defined as the number of nodes connected directly to the electrodes, e.g., $B = W + 1$ when all the elements along the boundary side are connected to the electrode. Each single node is represented by a corresponding linear algebraic equation, resulting in $(W + 1) \times (L + 1)$ equations for all the nodes. Two additional equations can be obtained from the electrodes. By solving these equations, the potential of each node and the current going through each bonds in the network can be calculated. Thus, for a given applied potential difference between electrodes, $|V_0|e^{i\omega t}$, the macroscopic admittance can be given by $Y = |I_0|e^{i(\omega t + \varphi_I)} / |V_0|e^{i\omega t}$, where $|I_0|e^{i(\omega t + \varphi_I)}$ is the obtained overall current flowing into the ground. Additionally, Frank-Lobb techniques can be employed to reduce the network size, thus improving the computational efficiency [31].

Here, the network aspect ratio and the electrode dimension are considered as variables, in order to investigate the influence of network configuration and boundary conditions on macroscopic responses. The network size is determined by its width, W , and length, L , which represent the numbers of components in a single chain along the horizontal and vertical directions, respectively. In the network circuit, two electrodes of identical dimension are connected to elements located symmetrically in the center of the two vertical boundaries. The frequency-dependent macroscopic responses obtained from different configurations, in terms of network length, L , and width, W , are normalized through

$$\tilde{Y} = \frac{|Y|RL}{W + 1}, \quad \tilde{\omega} = \omega RC, \quad (2)$$

where R and C are the resistance and capacitance values of resistors and capacitors in the considered network, respectively. This normalization process is applied in order to include the significance of all the elements in the overall network behavior, represented by the equivalent admittance, by considering rules for simple series and parallel combinations of components.

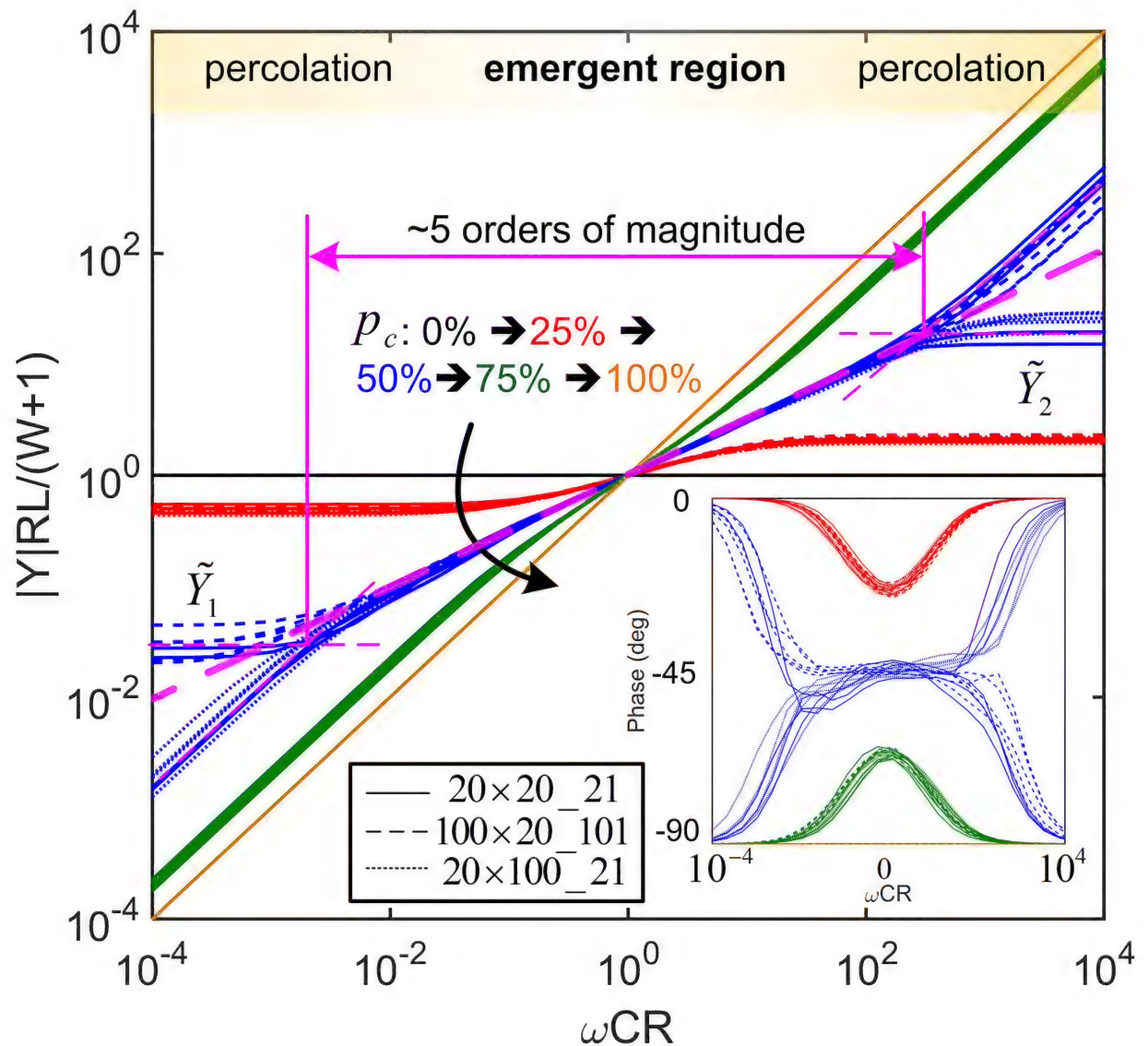


Fig 2. Normalised admittance module as a function of frequency. Numerical results obtained from three groups of differently configured networks (denoted as $W \times L_B$) are presented, with capacitor proportions, p_c , varying from 0% (corresponding curves are shown in black) to, 25% (red), 50% (blue), 75% (green), and to 100% (brown). The phase responses are depicted in the inset. For each network configuration with a given capacitor proportion, five simulations have been realized.

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The span of the emergent region, S , is defined as the horizontal distance between the intersections of the power-law function, $y = \omega^{p_c}$, with top and bottom percolation admittances, $\tilde{Y}_1$ and $\tilde{Y}_2$ (averaged from multiple simulations), for low and high frequencies, respectively, as shown in Fig 2.

The network behaviors shown in Fig 2 in the frequency domain are primarily governed by percolation effects [32], which are closely linked to the frequency-dependent conductivity of each single bond in the network. In the studied rectangular RC networks with two types of bonds (i.e., resistor and capacitor, the admittance ratio of capacitor elements with respect to resistors is $i\omega RC$) have been considered to describe the responses of random binary networks. The observed universal scaling behavior in Fig 2 can be also found in other networks

containing two types of elements, indexed by a and b , exhibiting differences which can be described in the form of $S_a = \omega^* S_b$, e.g., mechanical stiffness, thermal conductivity, and chemical reaction rate, etc. [11, 14, 25].

Results and analysis

The AC electrical responses of three groups of different sized networks (expressed in the form of $W \times L_B$, representing network width $\times$ length_electrode dimension: $20 \times 20_{21}$, $100 \times 20_{101}$, and $20 \times 100_{21}$) with varying p_c are plotted in Fig 2. The three types of regions observed in the conductance spectroscopy of square networks can be also observed here in rectangular ones. For a given network geometry, the obtained admittance spectroscopies for various p_c , are found to intersect at a convergence point with the characteristic frequency of $\omega = 1/RC$ where resistors and capacitors contribute equally to the overall conduction. This point also appears to be the center of the emergent region. The normalized characteristic admittance (the values of $\tilde{Y}$ at the characteristic frequency) at the convergence point is close to 1. This indicates that the network at the characteristic frequency perform effectively as a mono-element network, which has a phase angle reaching the extremum value, as is shown in Fig 2. It is further evidenced that the normalized emergent regions of the three groups of networks coincide with each other presenting universal features. The normalized admittance in this common emergent region appears to be uninfluenced by the network aspect ratio (length/width) or the electrode dimension. However, differences can be found at percolation regions along with the corresponding transition regions. Variation of the width or length can potentially change the percolation thresholds which will determine the responses, following a resistive-percolated (plateaued) or capacitive-percolated (upwards or downwards) trend, corresponding to the low and high frequency ranges, respectively.

Statistical analysis of the normalised characteristic admittance for different-sized square networks (from 5×5 to 600×600) with $B = W + 1$ was conducted and the standard deviation (STD) of the normalized characteristic admittance is presented in Fig 3. It is found that all values of normalized characteristic admittance obtained with various p_c (averaging over ten simulations) are in the range of (0.95, 1.05) for networks with more than 10×10 elements. The variance tends to diminish as the network size increases. This can be explained by considering boundary effects that relatively smaller networks have higher percentage of boundary elements (connected to four other elements rather than six in the bulk region). Responses of larger networks perform with little influence from the boundary. For a given sized square network a larger variation is found for cases of $p = 1/2$, as such conditions lead to an equal likelihood of resistive-percolated and capacitive-percolated network responses at low and high frequencies. Consequently, there are four possible qualitatively different types of response for any realization of the system.[21] Different available responses potentially introduce dispersion and uncertainty of the network behavior in both percolation and emergent regions, as can be seen in Fig 3.

We consider 2D rectangular networks with various L/W and $B/(W + 1)$ ratios, in order to study the effects of network size and electrode dimension on frequency dependent responses. The variations of normalized characteristic admittance for rectangular networks (not shown) are comparable to those of square networks. Here, the convergence-divergence behavior is tested with three groups of rectangular networks, which have the fixed width, W , of 20, 50, or 100 elements, respectively. The results obtained from the three groups coincide with each other, and typical results for $L/W = 1.5, 0.8, 0.2$ are shown in the Fig 4A. The contour of normalized characteristic admittance shown in Fig 4B presents a clear trend approaching 1, as the network length and the electrode dimension increase. For an RC network with a given size, a

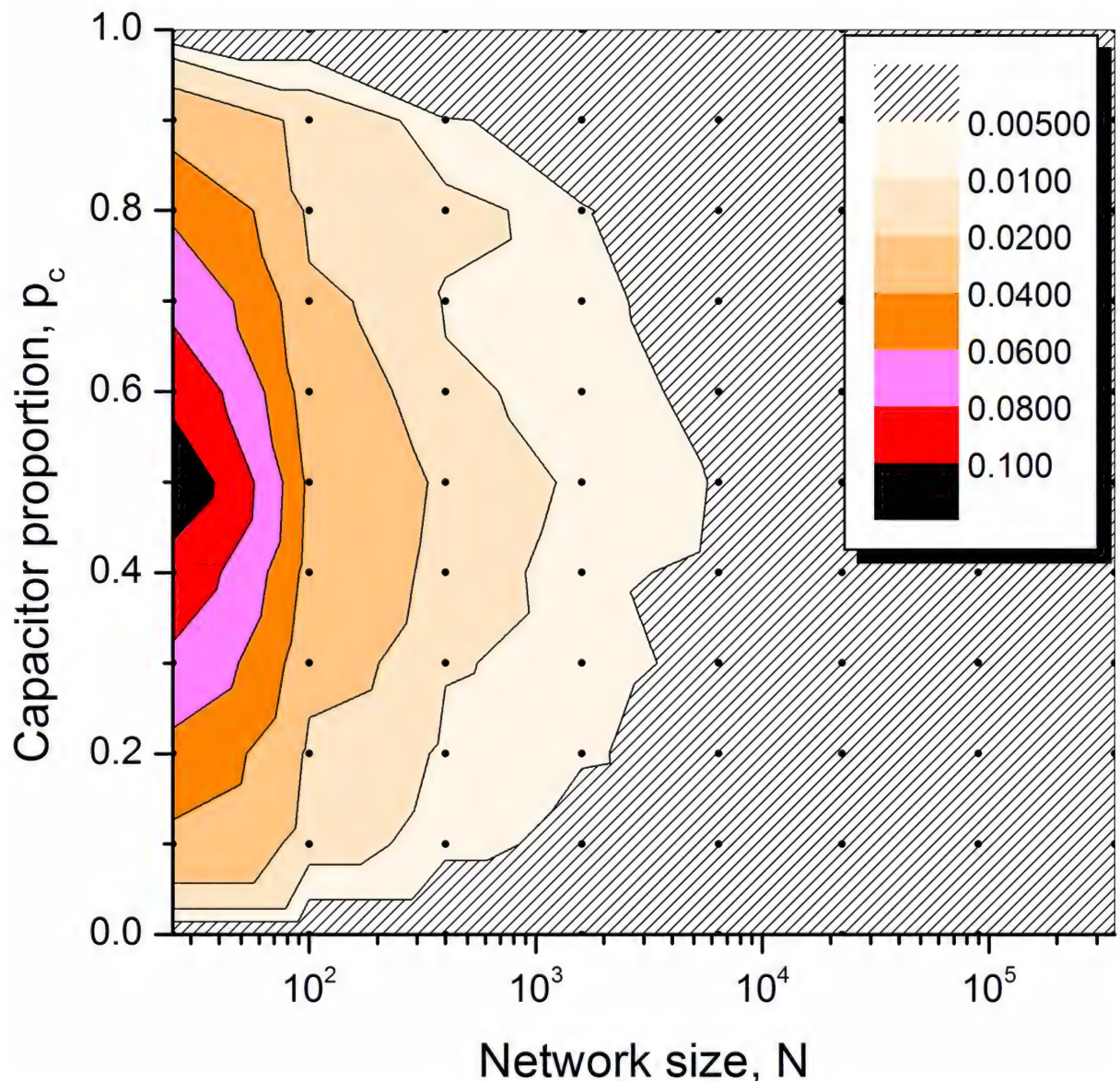


Fig 3. Mapping of the standard deviation of normalised characteristic admittance values. For varying capacitor proportions from 0.1 to 1.0, different-sized square networks (from 5×5 to 600×600) were considered. For a given network size and capacitor proportion, ten RC networks were generated and used in the simulations to obtain the averaged normalised characteristic admittance, represented by the black dot. The STD of these points are used for mapping with the colour indicating the STD values, as detailed in the legend.

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smaller value of electrode dimension tends to constrict the current to fewer paths at zones near the electrodes, thus, effectively reducing the network length. However, this influence will diminish as the network lengthens. Networks with large enough length tend to share the same intersection point uninfluenced by the boundary condition. In this case, these networks can be defined as homogeneous systems, with the responses in emergent region unaffected by the network configuration and the electrode dimension. A smaller length/width ratio or electrode dimension will usually lead to normalized characteristic admittance values smaller than 1.

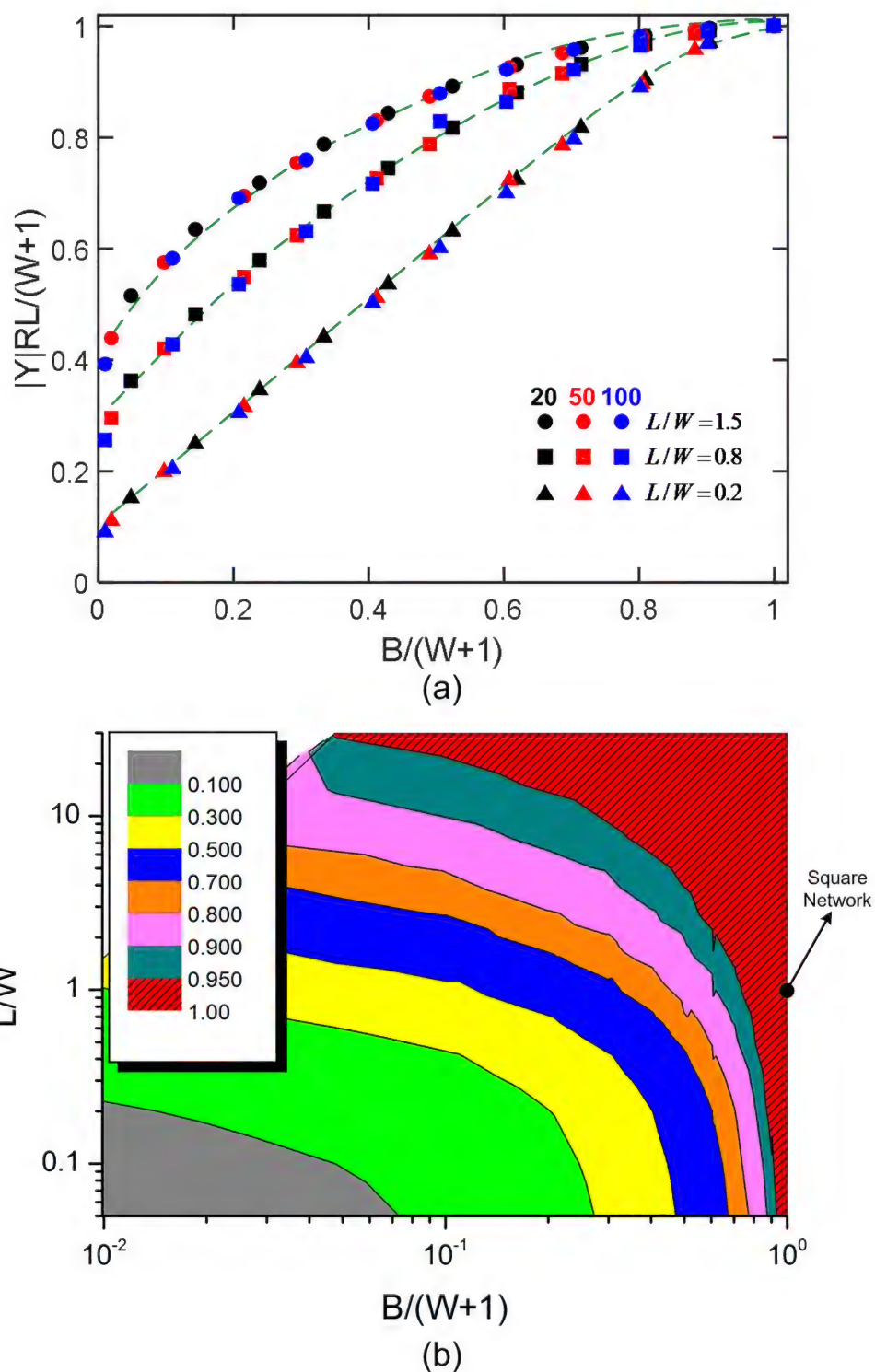


Fig 4. Influences of network aspect ratio and electrode dimension on the values of characteristic admittance. (A) The dependence of characteristic admittance on the $B/(W+1)$ ratio, for cases of different widths, W , of 20, 50, and 100 but same L/W , including 0.2, 0.8, and 1.5 (corresponding to $\blacktriangle$, $\blacksquare$, and $\bullet$, respectively). (B) Mapping of characteristic admittance values obtained from different-sized networks with varying L/W and $B/(W+1)$ ratios. The square network is marked by the black dot.

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For cases of electrodes of various geometries on the boundary or embedded in RC networks, the influence of the electrodes is mainly induced by the elements connected directly to the electrodes. As the network size increases, boundary elements and electrode-affected elements will have a decreasing percentage. Therefore, the electrode size effect along with the boundary effect will be unobservable for a sufficiently large network. The trend can be found in Fig 4B that the characteristic admittance values for an increasingly large network locate well upwards and to the right from the red zone, asymptotically approaching 1. This universal characteristic can be also extended for an infinite network, with the normalized intersection value to be 1.

The power law dependence of the electrical responses on frequency is of a universal nature for a wide range of complex RC networks. To further interpret this emergent behavior, we investigate the spans of the power-law emergent region. The convergence point tends to be the geometric center of all the emergent regions for various p_c . By considering the region center reported here combined with the span, S , the emergent scaling behavior region can be well described.

It has been observed [21] that, for a square network with $p_c = 0.5$, the span of the power-law emergent region increases without bound as the network size increases. In this paper, using the results from square networks as references, we compare the spans of emergent regions obtained for various sized rectangular networks with different electrode dimensions, as p_c varies. We found that when the normalized characteristic admittance value is sufficiently close to 1, it is the smaller dimension, $S_{min} = \min(W, L)$ that determines the span of emergent region, while the electrode dimension has little influence on the length. Different sized homogeneous networks with the same s_{min} tend to effectively present identical responses for the emergent region with a given p_c . However, discrepancies can be found for responses at low and high frequencies dominated by percolation behavior. This likely to be the case also for an intersection value far away from 1, but with lower accuracy due to the instability of the responses on account of the boundary effects. Only the results with p_c from 0 to 0.5 are discussed and presented in Fig 5, with the consideration of symmetricity of network responses.

The results shown here for varied network and electrode dimensions shed light on the behavior of infinitely-large binary percolation-type network and large networks with irregular boundaries (e.g., in the shape of spline curves) and electrodes (e.g., with the geometry of circular zones, embedded in the network, or unequal-sized electrodes). As long as the network size is significantly larger than the electrode and boundary dimension, the presenting universal features will not be influenced by the boundary and electrode conditions. This enables the network responses to reach a robust and reliable status at the emergent region with the span being determined by the effective network size in the order of D_e^2 (D_e is the distance between positive electrode and ground, indicating the shortest current path) and p_c (dominating the slope of universal power law). Additionally, evident trend presented in Fig 5 supports that infinitely large emergent scaling regions can be observed for various capacitor proportions.

Conclusion

We studied the influences of network geometry and electrode dimension on the electrical responses of rectangular RC networks. The universal scaling behavior can be fully characterized using the center and the length of the emergent region, i.e., the convergence point, and the span, respectively. For both square and rectangular networks, a convergence point is observed at the characteristic frequency, $\omega = 1/RC$, which usually appears to be the center of the emergent region. At this characteristic frequency, the normalized admittance value $|Y|RL/(W + 1)$ approaches 1 as the length-to-width ratio and electrode dimension increase. For a

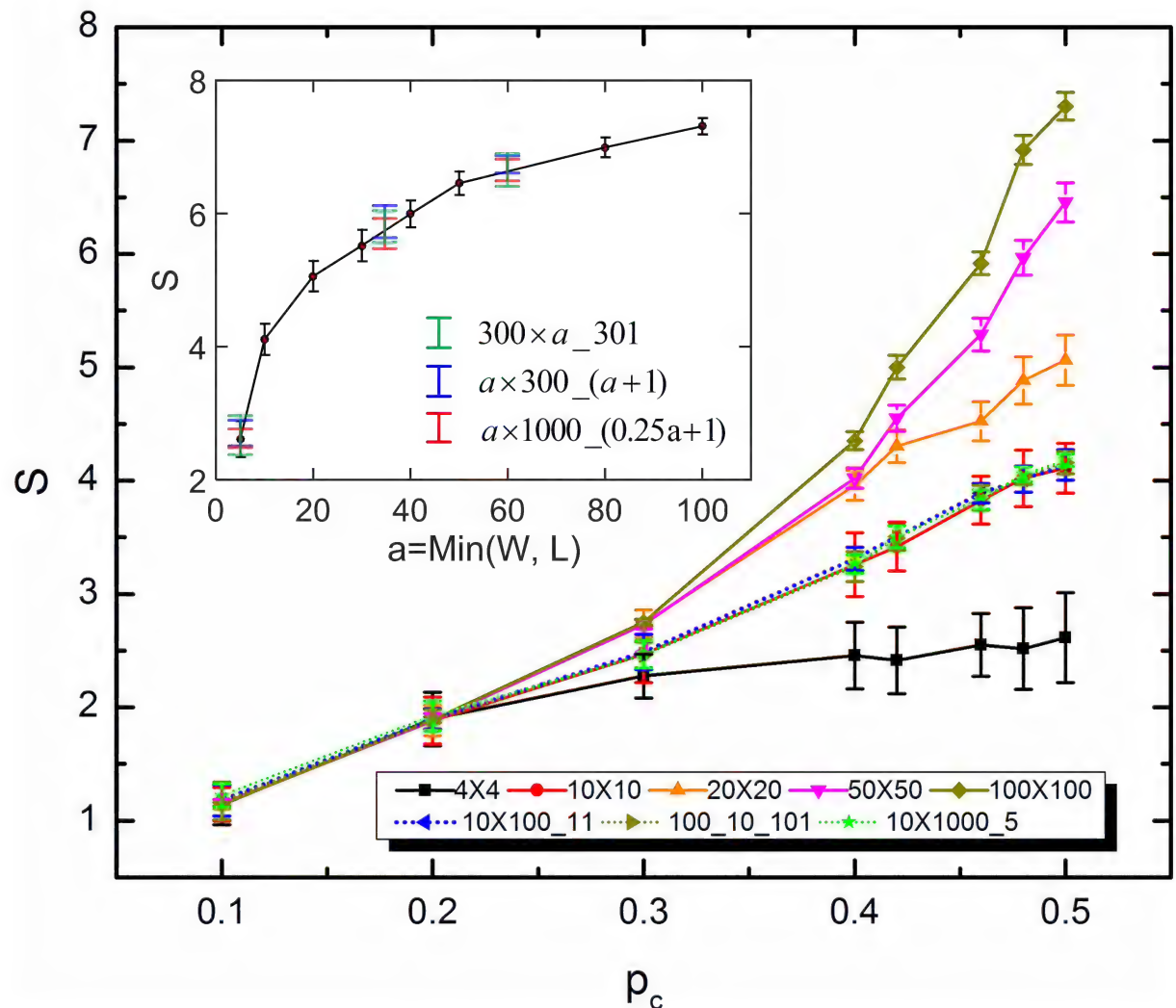


Fig 5. Dependence of the emergent region span on network size and electrode dimension. The results for square networks are shown by solid lines with error bars obtained across ten simulations for each point. Results of rectangular networks (10 × 100_11, 100 × 10_101 and 10 × 1000_5) are presented by dashed lines. The inset compares the spans of different-sized square networks with $p_c = 0.5$ with those of rectangular networks with various network configurations.

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defined homogeneous network, the span of the emergent range is primarily determined by the shorter dimension of width and length. These observations provide a unified description for the emergent scaling properties of network responses for random two-phase systems with varying topological configurations. The comprehensive understanding of this emergent scaling can guide the design and testing of disordered systems in terms of determining testing conditions (e.g., the shape, size, location, and spacing of the fixtures), boundary conditions, and system dimensions.

Author Contributions

Conceptualization: CZ DH YG.

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Formal analysis: CZ DH YG.

Funding acquisition: YG.

Investigation: CZ DH YG.

Methodology: CZ DH YG.

Project administration: YG.

Resources: YG.

Software: CZ.

Supervision: DH YG.

Validation: CZ YG.

Visualization: CZ YG.

Writing – original draft: CZ DH YG.

Writing – review & editing: CZ DH YG.

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6 Electrical Responses of Granular Materials

In this chapter, we extend the studies of electrical conductance in granular materials under DC scenario to AC scenario over a wide range of frequencies. This chapter containing Manuscript 6 presents experimentally observed universal AC scaling behaviour in conductive granular systems under different stress conditions. The spectra of impedance moduli on frequency follow a unique master curve with characteristic frequencies corresponding to the onset of conductance dispersion and measured direct-current resistance as scaling parameters demonstrating stress-independent universality. Three-dimensional RC network topographies are extracted from the random granular packing realized by DEM simulation. The information collected at inter-particle contacts are scaled up to simulate the macroscopic responses of the granular packing through the formed network structures. The inherent electrical responses are found to be governed by the network topology and inter-particle contact morphologies formed under a given stress level, by means of introduced numerical model based on 3D packing structures and equivalent RC networks. In Chapter 6, we break the electrical transport in granular materials down into electrical transport at individual inter-particle contact, in order to utilise the knowledge of contact mechanics presented in Chapter 3 and 4. Subsequently, based on the studies shown in Chapter 5, the information at inter-particle contact level is integrated upwards to macroscale through 2D/3D regular/irregular RC network structures to reveal the physical origin of the AC responses of granular materials.

I was the primary researcher and author of this manuscript, being supervised by Dr. Yixiang Gan, Dr. Dorian Hanaor.

Stress-dependent electrical transport and its universal scaling in granular materials

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Abstract

We experimentally and numerically examine stress-dependent electrical transport in granular materials to elucidate the origins of their universal dielectric response. The ac responses of granular systems under varied compressive loadings consistently exhibit a transition from a resistive plateau at low frequencies to a state of nearly constant loss at high frequencies. By using characteristic frequencies corresponding to the onset of conductance dispersion and measured direct-current resistance as scaling parameters to normalize the measured impedance, results of the spectra under different stress states collapse onto a single master curve, revealing well-defined stress-independent universality. In order to model this electrical transport, a contact network is constructed on the basis of prescribed packing structures, which is then used to establish a resistor-capacitor network by considering interactions between individual particles. In this model the frequency-dependent network response meaningfully reproduces the experimentally observed master curve exhibited by granular materials under various normal stress levels indicating this universal scaling behaviour is found to be governed by i) interfacial properties between grains and ii) the network configuration. The findings suggest the necessity of considering contact morphologies and packing structures in modelling electrical responses using network-based approaches.

The power-law scaling of dielectric properties with frequency known as the ‘universal dielectric response’ (UDR) [1] has been observed in a wide range of materials, including disordered ceramics [2,3], ion/electron-conducting glasses [3-5], amorphous semiconductors [4,6], metal powder [7] and nanoparticles [8,9]. This frequency-dependent behaviour is of importance in diverse applications including material characterization [2,4], battery optimisation [10], and electronic sensors [11]. Investigations of scaling in ac (alternating current) conduction have been conducted with square random resistor-capacitor (RC) networks [7,12-14], random-barrier models [4,15,16] and effective-medium approximation [16-18]. For different types of ionic [4,19] and electronic [9] conduction, the dependence of conductivity (σ) on frequency (ω) under a wide range of temperature conditions demonstrates a single master curve, suggesting the validity of the time-temperature superposition principle (TTS) described by the scaling law

$$\sigma(\omega)/\sigma^* = f(\omega/\omega^*), \quad (1)$$

where f is a temperature-independent scaling function, σ^* the dc (direct current) conductivity and ω^* the characteristic angular frequency corresponding to the onset of conductivity dispersion. This scaling behaviour may stem from various transport mechanisms in terms of timescales [4,5,15]. At different temperatures, T , all characteristic times, $\tau^* = 1/\omega^* \propto \exp(T^{-1})$, exhibit the same rate of ion hopping associated with activation energy, indicating the scaling of dynamic processes is temperature-independent [6,15,20,21]. Additionally, the proportionality of $\sigma^* \propto \omega^* \propto 1/\tau^*$ can be derived through the Barton-Nakajima-Namikawa relation, revealing TTS behaviour [22]. Such TTS is also observed for granular nanocomposites [8,9,23], where thermally-activated electron tunnelling among nanoparticles dominate electronic conduction, with $\tau^* \propto \sigma^* \propto \exp(T^{-1/2})$. In both ionic and electronic conduction τ^* and σ^* exhibit Arrhenius-type dependence on T^n [4,21], giving rise to the inherent spectral features with shape of the impedance spectra unaffected by temperature. The corresponding master curves for various materials are found to exhibit power law regions with exponents ranging from 0.6 to 1 within a certain frequency range [1,4,5,9,15,24,25]. At high frequencies, a regime of nearly constant loss (NCL) with the dispersion exponent approaching unity can be observed [15,26,27].

Scaling behaviour described by TTS can be expected with the fulfilment of $\tau^* \propto \sigma^* \propto \exp(\delta^n)$, where δ denotes the mechanical loading. Stress-dependent scaling in granular materials has seldom been studied and the mechanisms involved are not well understood. Moreover, few studies have focused on the underlying reasons for shape deviation in the TTS master curve [4,9,25]. In this letter, we examine the frequency-dependent electrical responses of randomly packed granular materials under various compressive loads. By considering inter-granular electrical transport within networks established from prescribed packing structures, we elucidate the fundamental drives shaping the master curve for various topological systems.

Monosized stainless steel spheres (AISI 304, with precision grade G200) were randomly packed in a nonconductive cylinder (Al_2O_3) of 85 mm in height, H , 98mm in diameter, D , with circular steel plates as top and bottom electrodes, as shown in Fig. 1. Impedance spectra were measured using an impedance analyser (Agilent 4294A) from 40 Hz to 10 MHz, and plotted as a function of frequency ω as shown in Fig. 2. We performed isobaric measurements for spheres of three different diameters (d), 1 mm (with applied pressures up to 434.43 kPa), 5 mm and 10 mm (1.91 kPa to 33.71 kPa). Within the applied

pressure range, all obtained impedance values are considerably larger than the total resistance ($< 10 \text{ m}\Omega$) of the measurement system excluding the packed bed under testing. In order to minimize current-induced local welding, we applied an ac signal with a peak-to-peak value of 200 mV, through which linear ohmic behaviour can be observed in current-voltage responses for the considered loading range [7,28,29], details can be found in Supplementary Material. Controlled measurements were performed to exclude parasitic resonance signals from cabling and connections, using measured Nyquist plots [30]. The measured impedance is characterised by a transition above a critical (angular) frequency ω_c from a low-frequency dc plateau to a dispersive region. At high frequencies, the impedance angle rises from negative to positive values under increasing mechanical load, indicating a capacitive-to-inductive phase transition. The studied systems can be simplified as networks formed by a large number of electrical components [3,12,13,31].

Here, an electrical network is established by incorporating electrical interactions between spheres. Adjacent particles can be treated as a capacitor. The interaction between two contacting particles will evolve from a capacitor (X_T , due to the existing oxide layer [33]), to an effective resistor (R_T) in parallel with an effective capacitor (X_T), under increasing compression [7,29,34-37]. As shown in Fig. 1, the interface tends to exhibit multiscale geometries governing interfacial electro-mechanical phenomena [38,39]. For a single contacting spot (i), the convergence and divergence of current give rise to a constriction resistance R_{ci} , where electrons are transported in different mechanisms depending on contact size, e.g. via Sharvin, Wexler, and Maxwell conduction [32,40-43]. Additionally, tunnelling resistance R_{ti} , in series with R_{ci} , through the oxide layer and voids contributes significantly to the resistance at an established micro-contact [23,40]. At the contact level, the effective resistance incorporates numerous electrical micro-contacts at interacting asperities. Micro-voids at contact regions in which trapped air is present form micro-capacitors (with capacitance presented by C_i) in parallel [29,34,37]. The total electrical contact impedance Z_T is formulated by

$$Z_T = X_b + R_T // X_T \approx \left[\frac{1}{\sum_i 1/(R_{ci} + R_{ti})} \right] // \left(\frac{j}{\omega \sum_i C_i} \right), \quad (2)$$

where the operator “//” represents parallel connection, j the imaginary unit, and X_b the impedance of the bulk, the value of which is typically orders of magnitude smaller than that of R_T and X_T , for the considered frequency ($\ll \text{GHz}$) [37]. Noticeably, large zones of metal-to-metal contact can be achieved with sufficiently high pressures, establishing continuous metallic pathways through contacting grains, leading to inductive system behaviour. Based on the interfacial properties, the ac responses of assemblies of 1 mm spheres are investigated using an RC network containing C and $R//C$ components, since only capacitive dispersion was observed within the applied loading range. At higher inter-particle forces, results showing inductive properties can be similarly realised by RCL networks containing C and $R//C + L$, where the operator “+” represents series connection and L represents the inductance arising from strong force chains [44].

To further analyse the stress-dependent plateau values and characteristic frequency in Fig. 3, we first identify these quantities from the impedance spectrum. As is shown in the insert of Fig. 4 (a), the typical reactance, Z'' , spectra for a 1 mm bed under varied compression exhibits peaks, Z''^* , at characteristic frequencies, ω^* , marking the beginning of conduction dispersion. This characteristic frequency increases

with load while the corresponding reactance peak diminishes in magnitude. Importantly, all Z'' curves follow a single master curve using Z''^* and the corresponding ω^* values as scaling factors, suggesting relaxation processes is stress-independent. At characteristic frequencies, the magnitudes of R_T^* and $X_T^* = 1/(\omega^* C_T^*)$, corresponding to representative values of a single interaction between a pair of spheres in the system, are similar [9,45], thus we have $\omega^* = 1/(C_T^* R_T^*)$.

The dependence of normalised plateau values, $\overline{R^*}$, on normalised applied pressure, $\overline{F^*}$, is presented in Fig. 3. The measured dc impedance of granular packing, R^* , obtained by averaging over ten tests and shown with error bars. The representative inter-particle force, F_T^* , and R_T^* , are linearly correlated to dimensionless load and resistance, respectively:

$$\overline{F^*} = \frac{4d^2(F_0 + F)}{(\pi D^4 E)}, \quad \overline{R^*} = D^2(R - R_C)/(HdR_0), \quad (3)$$

where $R = R^* + R_C$ is the measured resistance of the whole system, R_C the total resistance of the cables and connectors, R_0 the theoretical resistance of the bulk material for an identical volume as that of the packed bed, E the Young's Modulus of the tested material (203 GPa), F_0 the sum of the half weight of the granular assembly along with the top electrode, and F the applied compressive force. The characteristic time, $\tau^* = 1/\omega^* = C_T^* R_T^* \propto C_T^* \overline{R^*}$, is depicted as a function of $\overline{F^*}$ in Fig. 3. Straightforwardly, we can ascertain $\tau^* \propto \overline{R^*}$ due to the observed power law functions of $\overline{R^*} \propto \overline{F^*}^{-2.09 \pm 0.11}$ and $\tau^* \propto \overline{F^*}^{-2.01 \pm 0.10}$ with both exponents being approximately -2. The difference between the two exponents comes from C_T^* , which is affected by both the contact area and interfacial separation [29,34,37]. The absolute value of exponents for $\overline{R^*} \propto \overline{F^*}$ is found to decrease with applied load and sphere size, indicating that surface structure plays a diminishing role in conduction behaviour as inter-particle force increases. For 10 mm spheres, exponent values of 0.83 ± 0.14 and 0.32 ± 0.09 were found, corresponding to the five lowest and highest load levels, respectively. The latter is comparable to that (1/3) of Hertzian contact with Holm conduction as the dominant mechanism [28,41,43].

Finally, Fig. 4(b) shows $|Z|/R^*$ versus ω/ω^* for 1 mm grains with the load range of 1.91 kPa to 103.36 kPa, where Z''^* and ω^* can be recognized for the considered frequency range. $|Z|$ curves under all loading conditions collapse onto a single master curve with R^* and ω^* acting as scaling parameters, demonstrating inherent electrical features across a wide frequency range.

To explore the origins of the intrinsic electrical characteristics observed, we incorporated the electrical interaction between spheres into prescribed random packing structures realized using a discrete element method [46]. The obtained packing is sandwiched between two conductive rigid flats, one of which is grounded and the other is raised to a potential $V(\omega)$. The impedance can be obtained by solving the complex linear equations established by applying Kirchhoff's current law to each particle, denoting a node of the network [45]. A simulation volume larger than $10 \times 10 \times 10$ particle diameters has been recommended to reduce the size dependence of the percolation threshold [47,48]. Here, 8000 spheres were randomly packed in a rigid container with a height-area ratio and initial volumetric packing (62%). For comparison, increasing compression in experiments resulted in the volume ratio increasing from $61.233 \pm 0.859\%$ to $62.716 \pm 0.847\%$ over ten tests. With these settings, a coordination number N_c of

6.14 ± 0.11 over ten realisations is obtained, indicating the number of interacting spheres within the cut-off distance of a reference sphere [46,49,50]. This distance is initially set to equal the grain diameter.

For a 1 mm bed, either C or $R//C$ are assigned to “interacting” pairs of spheres. Consequently, the C element proportion (P_c^*) in a range $[0, 1]$ results in the total capacitor proportion (P_c) ranging from 0.5 to 1, as $P_c = 1/(2 - P_c^*)$. As a simplified assumption, all C and R elements are assigned identical values, C_s and R_s , respectively, across the network without referring to the contact force distribution. This assumption is justifiable on the basis of the robustness of RC network responses with respect to microstructural details and component positions [1,13,15,18,20] (see Supplementary Material). Moreover, the absolute value of the average contact force can be dimensionalised using Eq. (3). With this network structure, $R//C$ type components constitute the network backbone, providing equal numbers of capacitors and resistors. Spheres in weak contact, acting effectively as capacitors, add additional capacitive phases. Numerical and experimental results are compared in Fig. 4(b) with analyses based on a simple circuit-equivalence (a resistor and a capacitor in parallel) given for reference [7]. Higher values of P_c (smaller than the percolation threshold with $P_c \approx 0.83$ [51-53]) give rise to smoother transitions from resistive plateaus to NCL regimes. The master curve obtained from experimental results is well reproduced for $P_c = 0.70$. This P_c value is closely related to interfacial properties including roughness, oxide layer thickness and dominant conduction mechanism [7,28,32,38]. Moreover, the resistive plateau and onset of conduction dispersion are primarily dominated by P_c , as shown by the dependences of $|Z|/|R_s|$ and $\omega^* R_s C_s$ on P_c presented in the insert of Fig. 4(b).

To reproduce the observed inductive behaviour, RCL networks are simulated. As shown in Fig. 4(b), the onset of dispersion is found to be dominated by the dimensionless ratio, $\lambda = L/(CR^2)$, governed by interfacial properties [28,34,37]. Under increasing inter-particle forces, contacts evolve from C to $R//C$ and then to $R//C + L$ constituting a larger proportion, e.g., 0.9 used in the presented simulations. As a result, the current will percolate through inductors, and thus, present inductive dispersion at high frequencies [32].

We have endeavoured to interpret experimental measurements with a reasonable working hypothesis based on interfacial properties. In order to investigate the role of packing structures in the universal scaling characterized by the master curve, we further applied the presented numerical framework with $P_c = 0.70$ to networks with various configurations. Rather than 2D or 3D square lattice networks [12,15,18,45], we vary the cut-off distance defining the limits of particle interactions to obtain network structures with different coordination numbers [46,48,51]. Networks with mean N_c values of 4.02 ± 0.15 , 6.01 ± 0.20 , 7.99 ± 0.16 , 10.13 ± 0.21 , and 12.08 ± 0.16 over ten realizations of packings were achieved, corresponding to applied cut-off distances of $0.99 d$, $1.02 d$, $1.11 d$, $1.30 d$, and $1.45 d$, respectively. As is shown in the insert of Fig. 4(c), a higher N_c results in a sharper transition from resistive to NCL regimes, a later onset of conduction dispersion and a lower resistive plateau value. The coordination number indicating the connectivity of the network structure, is associated with the effective local dimension of current pathways, which fundamentally determines the conduction properties of network structures [51,52], and thus the transition from resistive plateau to dispersive conduction as frequency increases.

From the comparison above, it can be concluded not only P_c but also N_c is important in determining the shape of the impedance vs frequency master curve of ac electrical transport in granular materials under various stress states. Conventional models generally comprise ordered 2D or 3D square lattices [18,54,55] with constant particle coordination, and furthermore do not incorporate the electrical conduction mechanisms intrinsic to granular materials. Consequently, such analyses tend not to reproduce the impedance-frequency scaling of granular systems with various packing structures, and limit their utility towards the precise interpretation of observed transport phenomena. The present results demonstrate that by considering the topological configurations of conductive granular systems along with the physical characteristics of intergranular contacts, ac scaling behaviour in these systems can be meaningfully reproduced. The proposed framework sheds light on transport analyses of both electrons and ions in discrete materials and systems at various length scales, e.g., percolation-dominated conductivity [45,48,50,52,56], origins of NCL [4,12,57], dependence of ac electron/ion conductance curve on dimensionality [4,5,15,25], etc.

We have presented here experimental observations of universal ac scaling behaviour in conductive granular systems under different stress states. Results reveal that impedance moduli curves collapse onto a unique master curve with R^* and ω^* serving as scaling parameters, proving stress-independent universality. By validating our experimental results with the introduced numerical model based on 3D packing structures and equivalent RC networks, we have shown how both this scaling and the observed capacitive-to-resistive transition emerge as a consequence of percolation, which in turn is governed by network topology and inter-particle contact morphologies formed under a given stress level. Overall conduction in applied granular systems is thus fundamentally driven by the manner in which individual particles interact with each other in compression to give rise to resistive and capacitive elements and collectively produce percolation pathways. It flows from this that the mechanical and topological features of applied granular systems are key towards understanding their electrical responses.

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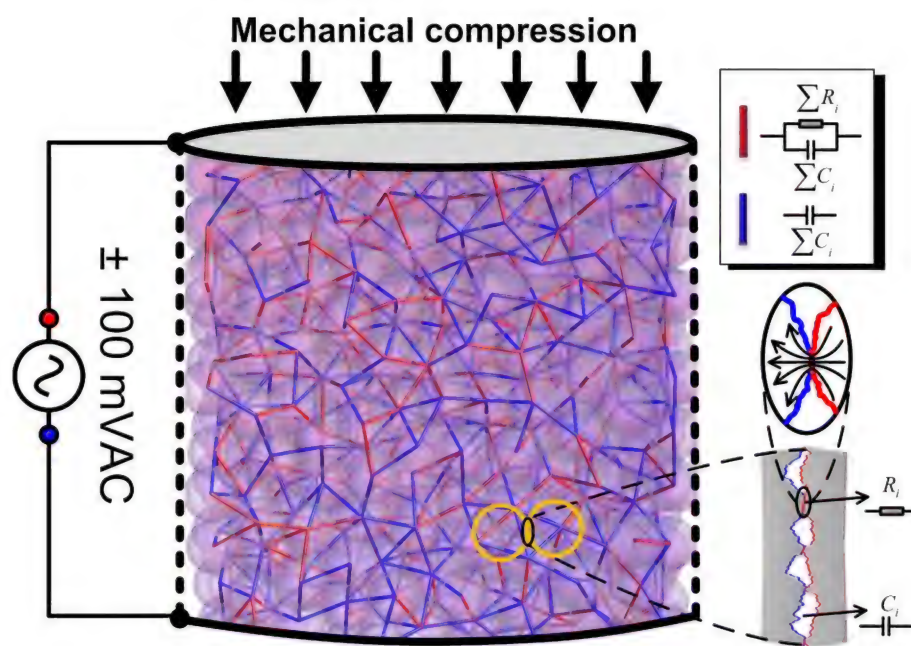


FIG. 1. Schematic of the electrical conduction in granular materials. The established RC network structure is presented with red and blue bars, respectively, indicating capacitors and parallel resistor-capacitor units.

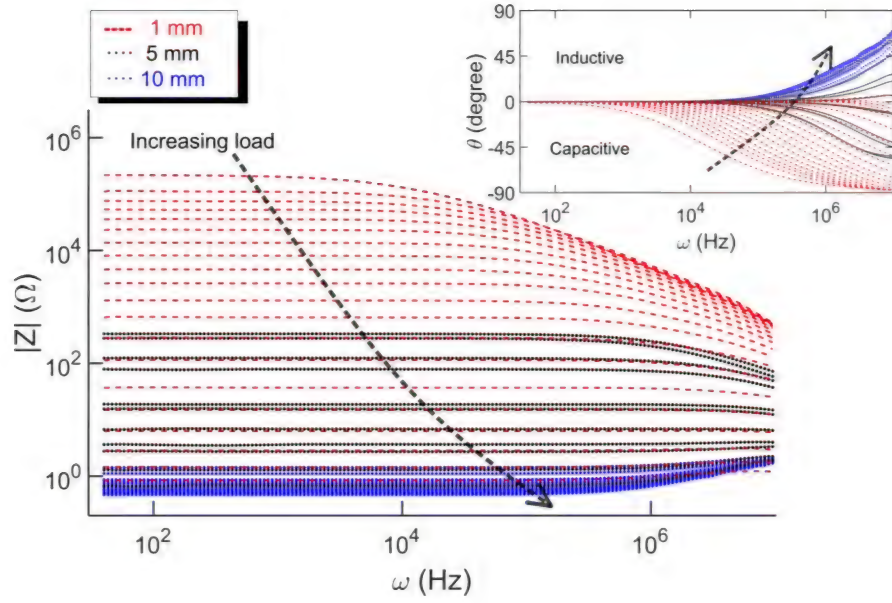


FIG. 2. Typical measured impedance module, $|Z|$, as a function of frequency, ω . Insert shows the impedance phase, θ , versus frequency. Results for spheres of three different sizes are presented using red, black and blue lines, corresponding to, 1 mm, 5 mm and 10 mm diameter spheres, respectively.

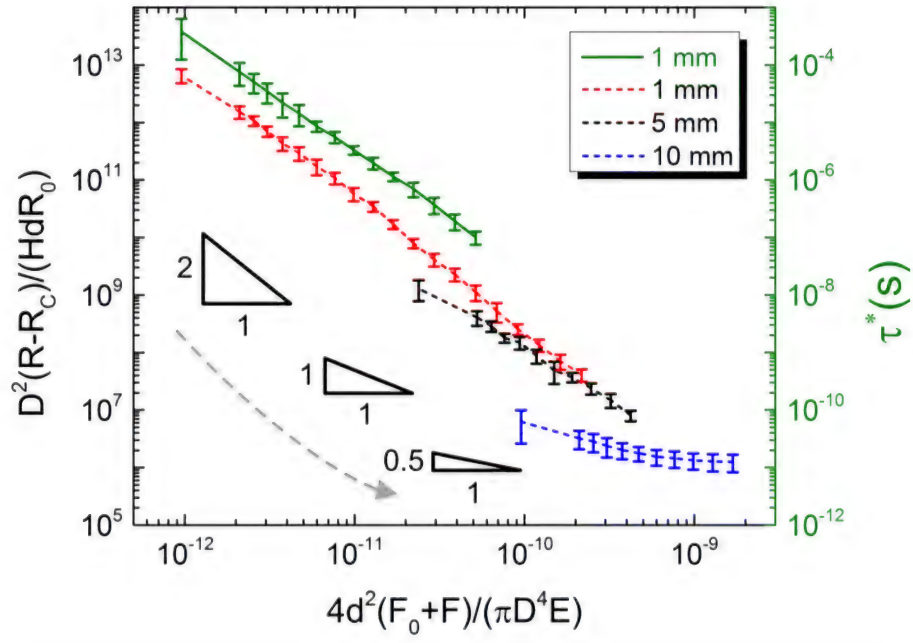


FIG. 3. Dependence of dimensionless $\bar{R}^*$ on $\bar{F}^*$ for assemblies of different-sized spheres shown with dashed lines with error bars. The dependence of τ^* on $\bar{F}^*$ for 1 mm diameter spheres, with pressures from 1.91 kPa to 103.36 kPa, is presented in solid line corresponding to the right y-axis.

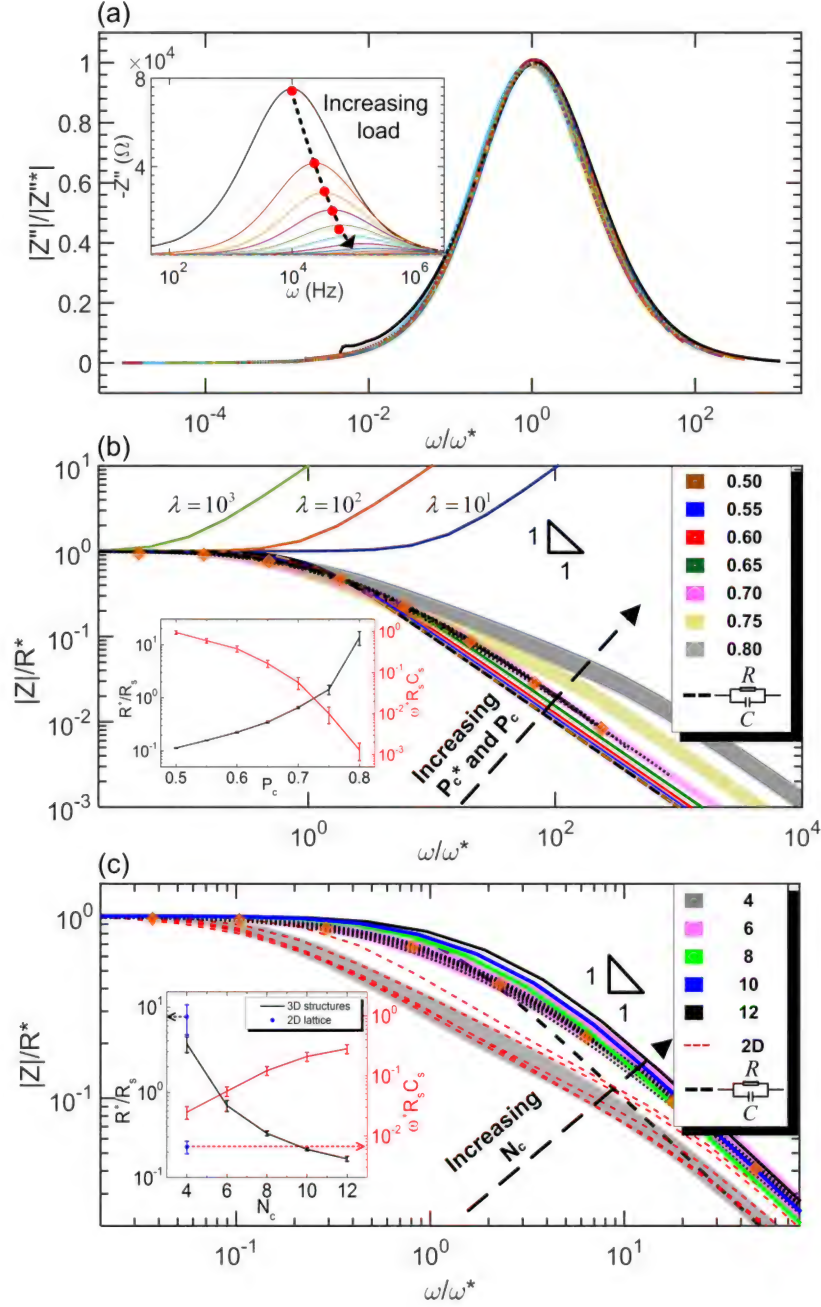


FIG. 4. (a) Typical scaling of $|Z''|/|Z'''|$ versus ω/ω^* for 1 mm bed. Insert shows Z'' as a function of frequency with red dots marking Z''' at ω^* . (b) Scaling plots of $|Z|/R^*$ versus ω/ω^* . Experimental results for 1 mm diameter spheres are shown in black dots with the fitted master curve marked with orange diamonds. Simulation results of RC and RCL networks, respectively, with various P_c and λ presented by shaded error bars. (c) Simulation results based on random packings with N_c from 4 to 12. Red dashed lines present typical master curves for 2D square lattice networks with R and C components. The inserts in (b) and (c) present R^*/R_s versus N_c and $\omega^*R_sC_s$ versus N_c , corresponding to the left and right y-axes, respectively. The ac response of simple R/C equivalence is presented by the black dashed line. Each individual point shown in error bar is based on ten realizations.

Supplemental Material: Stress-dependent electrical transport and its universal scaling in granular materials

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1. SYSTEM CALIBRATION USING DIRECT CURRENT

Sweep tests have been conducted in order to examine the influence of testing current levels on measured responses. Here, the conducted sweep test consisted of five successive back-and-forth scanning current cycles with increasing current range. Each applied cycle included four phases with the “loading” phase ① having a logarithmically increasing current, the “unloading” phase ② having a logarithmically decreasing current, followed by “loading” phase ③ and “unloading” phase ④ with reversed current direction. Typical obtained current-voltage curves for 1mm spheres are shown in Fig. 1. For the impedance spectroscopy presented in this study, we employed an ac voltage with a peak-to-peak value of 200 mV, with which a linear Ohmic behaviour can be observed in current-voltage responses for the considered loading range.

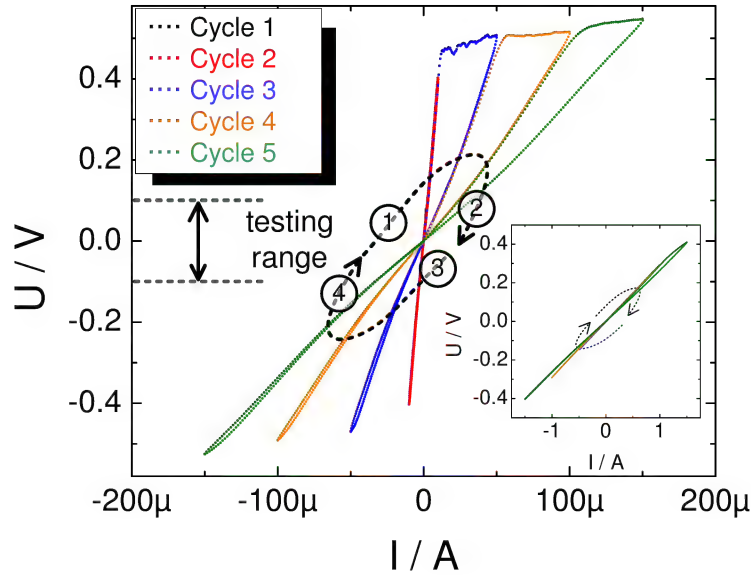


Fig. 1. Observed hysteresis curves for a bed of 1 mm spheres with five successive back-and-forth sweeping current cycles of increasing ranges, under a constant stress of 3.82 kPa. Insert show the recorded current-voltage responses under a stress of 509.39 kPa.

2. CHARACTERIZATION OF SPHERE ROUGHNESS

The surfaces of spheres were scanned using an optical surface profilometer (NanoMap 1000WLI) to obtain three-dimensional (3D) digitised topographies with 1024×1024 pixels. The apparent curvature corresponding to the sphere diameter is removed, in order to compare the surface geometry at finer scales which plays a dominant role in contact properties [1,2]. Fig. 2 shows typical surface structures from three different-sized spheres used in this study. These 3D surface structures were then characterized using roughness parameters including amplitude roughness, root mean square roughness, root mean square slope and fractal dimension using triangulation method [1], as is presented in Table 1. The shown standard deviations were calculated over ten scans on different samples. The three groups of different-sized spheres are found to exhibit similar surface structures, however, behave differently under a given inter-grain force owing to the various roughness-to-diameter ratios [3,4].

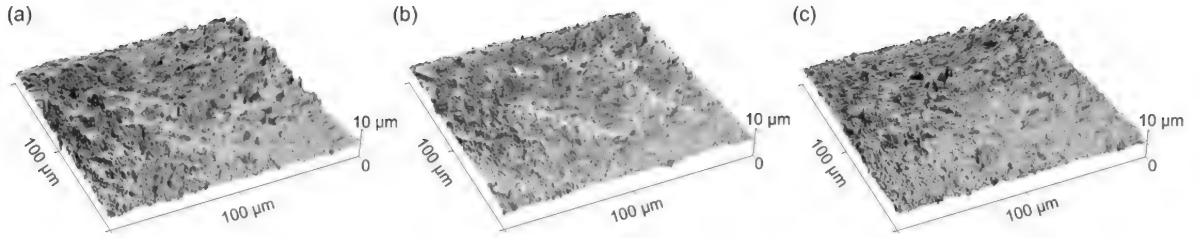


Fig. 2. Surface roughness for spheres with different diameters: (a) 10 mm; (b) 5 mm; (c) 1 mm.

Table 1. Surface characterisation of different-sized spheres

Sphere size	Amplitude roughness $R_t / \mu\text{m}$	Root mean square roughness $R_{rms} / \mu\text{m}$	Root mean square slope R_s	Fractal dimension D_{tri}
10 mm	7.026 ± 1.864	0.594 ± 0.218	0.071 ± 0.002	2.310 ± 0.035
5 mm	9.872 ± 2.232	0.303 ± 0.111	0.102 ± 0.006	2.323 ± 0.092
1 mm	9.147 ± 1.297	0.439 ± 0.164	0.089 ± 0.009	2.374 ± 0.073

3. ROBUSTNESS OF NETWORK RESPONSES TO MICROSTRUCTURES OF SPHERES

In order to further study the effects of components values arising from inter-grain properties on electrical responses, we conduct simulations with resistors and capacitors of various values. The distribution of resistors and capacitors values are achieved by multiplying the initially prescribed identical value a distribution factor. Using the proposed simulation framework presented in manuscript, we obtain the electrical responses of network containing C and R/C with the resistor and capacitor values following Gaussian and uniform distributions, as is shown in Fig. 3. The inserts show the distribution factors, U , and G , corresponding to the employed uniform and Gaussian distributions. Here, insensitivity of network responses to the values and positions of elements can be observed which has been also reported in previous literatures [5-8].

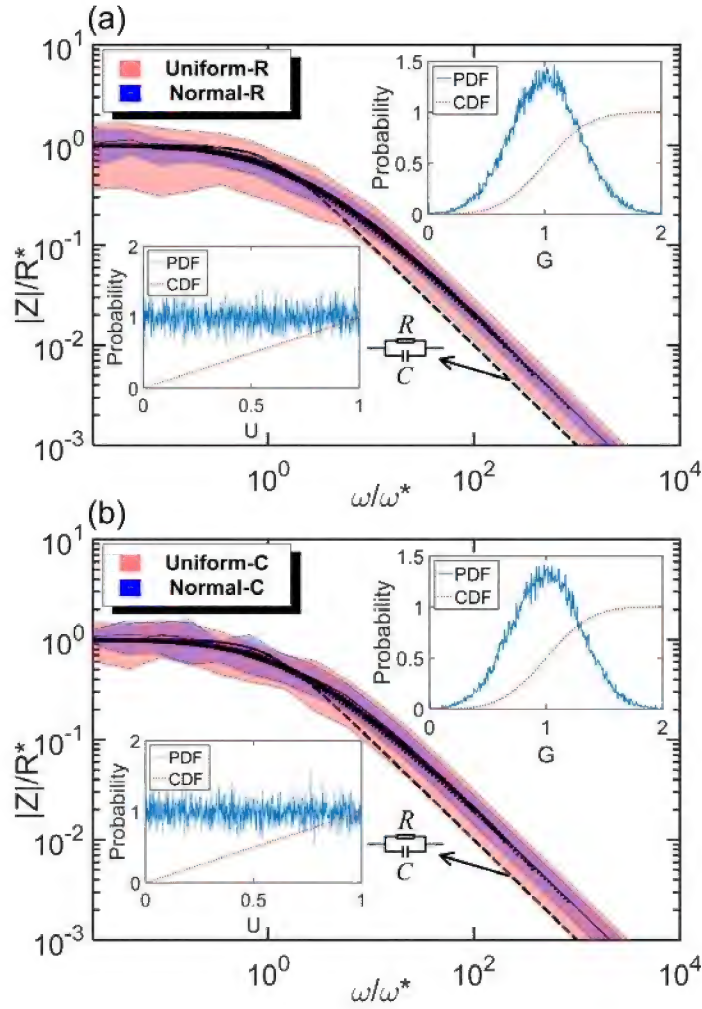


Fig. 3. Scaling of $|Z|/R^*$ versus ω/ω^* for networks containing C and $R//C$ with (a) resistors values and (b) capacitors values following uniform and normal distributions, respectively, corresponding to the red and blue shaded error bars obtained based on ten realizations. Inserts show PDF and CDF of the distributions of capacitor and resistor values. Experimental results for 1 mm diameter spheres are shown in black dots. The ac response of simple R/C equivalence is presented by the black dashed line.

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7 Conclusion

This work has introduced a physics-based framework combining features containing experimental and numerical information obtained across various length scales, thus providing a comprehensive model that can guide the design and optimization of the electrical conduction in granular systems. Major conclusions achieved in this thesis are summarized below:

- (1) The influences of surface roughness on the normal contact stiffness were experimentally and numerically studied. For both approaches, a power-law dependence of normal contact stiffness on normal compression has been observed. The exponent of this relationship has been demonstrated to be closely correlated with the fractal dimension of surfaces, while the amplitude depending on the root mean square roughness. These empirically derived strong correlations can be used for establishing and validating predictive models for properties of inter-particle contact.
- (2) The electrical contact resistance at interfaces, measured using a controlled current method, revealed that the measured resistance is affected by testing current, mechanical loading, and surface topology. With an applied current at proper level, the holmic electrical transport can be observed. Within the holmic scale, the electrical contact resistance as a function of applied normal pressure is found to follow a power law within a certain range, the exponent of which is closely linked to surface topology characterized by roughness amplitude and fractal dimension.
- (3) The correlation between stress-dependent electrical contact and normal contact stiffness is derived to show the intrinsic electro-mechanical coupling at rough interfaces. With the performed experimental and numerical investigations into the contact stiffness and electrical contact resistance at rough interfaces focusing on their dependence on applied force, we provide a first-order investigation connecting interfacial mechanical and electrical behaviour, applicable to studies of multi-contact systems.
- (4) The effects of network configuration on macroscopic network responses of two-dimensional finite binary percolation networks have been examined. For given network geometries, the normalised macroscopic frequency-dependent electrical conductivities for different capacitor proportions are found to converge at a characteristic frequency. Networks with sufficiently large size have been demonstrated to share the same convergence point uninfluenced by the boundary and electrode conditions. The span of the emergent power-law scaling region is found to be primarily determined by the smaller network dimension (width or length). This study identifies the power-law scaling in random two phase systems of different topological configurations, having implications on the design and testing of disordered systems in diverse applications.
- (5) The stress-dependent electrical transport in granular materials was experimentally studied. The macroscopic responses under alternating current with varied compressive loadings consistently exhibit a transition from a resistive plateau at low frequencies to a state of nearly constant loss at high frequencies. The spectra under different stress states collapse onto a single master curve by using characteristic frequencies corresponding to the onset of conductance dispersion and measured direct-current resistance as scaling parameters to

normalize the measured impedance. This universal response for various loading conditions reveals well-defined stress-independent universality.

- (6) The electrical conduction in granular materials has been simplified to an electrical network formed by a large number of RC components in random positions on a 3D basis. The experimentally observed master curve can be meaningfully reproduced. The overall conduction in applied granular systems is fundamentally driven by the pattern in which individual particles interact with each other to produce current percolation pathways, and the intergranular contacts determining resistive and capacitive elements.

Together, information collected across a wide range of length scales allows us to have access to a comprehensive model that is desirable for understanding the electrical conduction in granular systems. The contact properties of rough interfaces are extended downwards to the asperity level based on the fractality of surface geometries and integrated upwards to the macroscopic scale through network configuration. Finally, the combination of the studies across scales from asperities, interfaces, grains, to device scales leads to new constitutive models that can contain information collected from lower scales and is applicable to different categories of granular materials.

Future outlook

Clearly, there is much work to be done to push this work further. In the future, several directions seem to be important for further developments for the conduction behaviour in granular materials:

- (1) Firstly, this work, incorporating the properties arising from discrete nature of materials, can be further developed to provide a general continuum model applicable for electrical conduction in disordered systems with irregular-shaped particles described by anisotropy contact network and particles of various materials exhibiting different electrical properties. Moreover, the network framework introduced in this work can potentially be used to describe other transport phenomena, such as the AC ion-conductivity of amorphous materials, multiphase flow in porous media, biological systems, and hopping conduction in semiconductor and composite materials.
- (2) Many different numerical strategies were proposed in solving the contact problem between particles of granular materials, ranging from classic Hertzian contact, asperity-based models via Persson's theory and brute-force computational approaches, to molecular dynamics simulations, in which the original assignment was scaled down to the atomistic scale. Each group of approaches lead to satisfying answers for at least one of the posed questions in contact mechanics like adhesion, friction, contact stiffness, contact resistance, etc. However, efficiency, versatility, and accuracy differed between methods. The more precise methods being, in general, computationally more complex. It remains challenging to properly and efficiently select among the existing approaches including the numerical method proposed in Chapter 3 for inter-particle contact within granular systems.
- (3) The fractality of a rough surface has inspired numerous multi-scale approaches to quantitatively describe the rough contact. It is a critical problem in employing the fractal-based methods about the fractal limit and the sensitivity of cut-off wavelength defining the finest features of involved the surface structures. Additionally, extra efforts should be made to propose the model for inter-particle contact with the consideration of radius/roughness

ratio, evolution of surface structure during compression and cyclic loading, and electrical-thermal-mechanical coupling at asperities.

- (4) This study has implication in developing 3D topography based on the relationship between localized electrical properties and the force distribution in granular materials. The evolution of the inter-particle force under varying applied loads can be sensitively reflected from the electrical responses of localised network. Therefore, the inter-particles forces and the effective stress in granular materials can be obtained based on the measured electrical conductivity and estimated current pathways.
- (5) The electrical transport at rough interfaces and in granular materials under dynamic conditions are of particular interest and importance for many applications. This work will provide a basis for the contact and conduction problems in vibrating environments.

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